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Pre-Conference Abstracts

SYMPOSIUM ON THE TROPOSPHERIC CHEMISTRY OF THE ANTARCTIC REGION

A. Hogan and S.L. Bowen, Editors

June 1991

Cooperative Institute for Research
in Environmental Sciences
University of Colorado

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Special Report 91-10

This volume contains authors' abstracts, which were reviewed by the organizing committee and accepted for presentation in both poster and podium format. The abstracts are arranged alphabetically by first author. The session number is listed at the top of each abstract.

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Cooperative Institute for Research in
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University of Colorado
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June 3–6, 1991

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Special Report 91-10 June 1991

DEDICATION
Dr. William H. Zoller

Dr. Zoller in the meteorologists' Jamesway, South Pole, January 1975

We dedicate this volume to Dr. William H. Zoller. Bill initiated his aerosol program at the old South Pole Station. He overcame tremendous problems with respect to station pollution and pioneered the concept of a clean air sector at the South Pole, reserved for experiments which required an undisturbed look at the Antarctic environment. Every tropospheric chemistry program in Antarctica is indebted to his initial and continuing efforts to maintain an uncontaminated observatory.

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P2.15

LIDAR MONITORING OF AEROSOL AND CARBON DIOXIDE IN THE LOWER TROPOSPHERE

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At present, lidar techniques are successfully used to study tropospheric aerosols. Lidar systems help to understand dynamic processes in the atmosphere. This paper describes the lidar systems used to study the tropospheric aerosols and gas composition of the atmosphere, aerosol and humidity distribution in the atmospheric underlayer over the ocean and experimental measurements of CO_2 concentrations. Two wavelengths were used to sound aerosols, λ=532 nm and λ=1064 nm. Lidar systems installed on a research vessel helped to resolve aerosol concentration profiles up to 10 km and water vapor concentration profiles up to 1.5 km in height. Stationary coastal lidar ensured the height of aerosol sounding up to 30 km. In the atmosphere/ocean interlayer, the height distribution of aerosol concentrations and humidity fields for various meteorological situations has been studied. The data are obtained for the inverse humidity layers formed 100-300 m over the ocean which cause the formation of aerosol layers with greater concentration. The observations are carried out for the night convection of water vapor destroying inverse layers. The measurements of CO_2 concentration in a clean atmosphere proved possible to register changing CO_2 concentrations 100 m over the ocean at a distance up to 300 m.

3.2

MEASUREMENTS OF ATMOSPHERIC METHANE AT JAPANESE ANTARCTIC STATION, SYOWA

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Precise and continuous measurements of the atmospheric methane (CH_4) concentration have been initiated at Syowa Station (69°00'S, 39°35'E) in February 1988. The measurement system was developed using a gas chromatograph (GC) equipped with a flame ionization detector. Precision was typically ±0.07% in a range from 1000 to 2000 ppbv, which was attained by improving the gas flow lines and electric circuit of the commercially available GC. The standard gases were CH_4-in-air mixtures prepared gravimetrically with an extremely precise balance, and uncertainties in their CH_4 concentrations were estimated to be ±0.2%.

The CH_4 concentrations measured at the station were extremely stable, and outliers due to local contaminations such as station activities were hardly observable. Regular diurnal CH_4 variation was not seen throughout the year, but the seasonal variation and annual increase of the concentration were clearly observed. The average seasonal variation for 1988 and 1989 showed minimum and maximum concentrations early in March and late in September, respectively, and peak-to-peak amplitude of 30 ppbv. The annual mean values of the CH_4 concentration were 1640 and 1651 ppbv for 1988 and 1989, respectively, yielding the increase of 11 ppbv yr^{-1} for the two years. The result of this measurement suggests that the recent annual increase rate of the CH_4 concentration is smaller than previous reported values of about 1% yr^{-1}.

2.6

TRACE ELEMENTS AND INDIVIDUAL PARTICLE ANALYSIS OF FIVE YEARS OF AEROSOL SAMPLING IN THE ANTARCTIC PENINSULA

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The recent concern about the possibility of changes in the global atmospheric composition has pointed out the importance of long-term monitoring of gases and aerosol particles in remote sites. Atmospheric aerosols were sampled continuously since December 1985 at the Brazilian Antarctic Station "Comandante Ferraz," (62°05'S; 58°23.5'W) in the King George Island, Antarctic Peninsula. Stacked Filter Units (SFU) were used to collect fine ($d_p < 2.0 \mu m$) and coarse ($2.0\ \mu m < d_p < 15\ \mu m$) particles on Nuclepore filters. The concentration of elements with Z>10 were measured by particle-induced X-ray emission (PIXE) analysis. It measured the concentration for 23 elements: Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Se, Br, Rb, Sr, Zr and Pb. The fine and coarse aerosol mass concentration was determined by gravimetric analysis. Absolute principal factor analysis (APFA) was used to obtain the aerosol elemental source profiles. The major and trace elements show a strong seasonal variability. For summer and wintertime coarse particles, only two factors were significant, with the first having high loadings for Na, Mg, Cl, S, Sr, K, Ca, and the coarse particle mass concentration (representing sea-salt aerosol). The second factor had high loadings for Al, Si, Fe, Ti, and Ca (soil dust aerosol). For the fine mode, three factors were differentiated, and they represented sea-salt aerosol, soil dust and sulfates. The sea-salt source profile agreed with the average sea-water elemental composition to within 20% for the elements Na, Mg, S, Cl, K. Ca and Br. For the soil dust component, the elements Mg, Al, Si, K. Ca, Ti, Mn, and Fe were present to within 40% of the average bulk elemental composition for the earth crust. The source apportionment for the coarse particle mass concentration (CPM) reveals that 76% of the CPM is accounted for by the sea-salt aerosol component, 4.5% by soil dust and 20% of the CPM could not be apportioned. For the fine particle mass concentration (FPM), sea-salt aerosol accounted for 80%, sulfates for 16%, soil dust for only 1.1%, and 3.5% of the FPM could not be apportioned. Excess sulfate shows a clear seasonal pattern. The concentration of some trace elements like Cr, Ni, Cu, Zn, and Pb appear too high, maybe due to long-range transport or other anthropogenic sources in the Antarctic Peninsula. Individual particle analysis by Electron Probe X-Ray Microanalysis (EPMA) shows a variety of types of particles. Calcium sulfate particles show a strong seasonal variability, accounting for up to 8% of the number of particles during summertime. In wintertime, sulfur is present in several types of particles, appearing at concentrations of 1 to 5% in mass, and is only sporadically present as pure ammonium sulfate particles.

P2.4

TRACE ELEMENTS AND BLACK CARBON IN AEROSOLS FROM THE ANTARCTIC PENINSULA

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There are very few measurements of fine mode trace elements and black carbon in the Antarctic atmosphere, especially during wintertime. In order to fill this gap, atmospheric aerosols were sampled continuously from March to November 1989 at the Brazilian Antarctic Station "Comandante Ferraz" (62°05'S; 58°23.5'W) in the King George Island, Antarctic Peninsula. A special sampler to collect fine mode particles in a small area was used. A cyclone is employed to cut off large particles and a 25-mm diameter 0.4-µm Nuclepore filter collects the aerosol particles in a 0.8-cm^2 area. The flow rate was about 6 lpm and sampling time was seven days. The concentration of elements with Z>10 was measured by particle-induced X-ray emission (PIXE) analysis. It was possible to obtain concentrations for 20 elements: Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Mn, Fe, Ni, Se, Br, Rb, Sr, Zr and Pb. Black carbon was measured with a soot photometer, with blank values taken from the same Nuclepore filter where particles were collected. Marine aerosol particles are responsible for more than 95% of the aerosol mass. The concentrations of soil dust related elements (Al, Si, Ti, Mn, Fe) were very low. The average S concentration was 51 ± 62 ng/m^3, with strong seasonal variability, and with an average enrichment factor for marine aerosol of 2.3. Excess sulfur also shows strong seasonal variability, with very low values during wintertime. The average black carbon concentration was 2.2 ± 1.2 ng/m^3, with minimum concentration of 0.7 ng/m^3 and maximum concentration of 4.7 ng/m^3. These values are very low, but they should be interpreted as upper limit concentrations, since some contribution from ships and diesel power generators can contribute to black carbon concentration. There was no correlation of black carbon with trace element concentrations. Enrichment factor calculations show that Na, Mg, Cl, K, Ca, Br, and Sr are sea-salt related, with EF values between 0.7 and 1.8. Al, Si, Ti, Mn and Fe have EF from 0.8 to 1.6 if average crustal rock is used as reference source. Factor analysis shows four components: sea-salt, soil dust, sulfates and black carbon. Sulfur is apportioned to both sea-salt and sulfate aerosol components.

P5A.8

SENSITIVITY OF AIR PARCEL TRAJECTORIES COMPUTATION OVER THE SOUTHERN HEMISPHERE TO THE UNCERTAINTY IN THE DETERMINATION OF THE WIND FIELD

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This paper studies the sensitivity of low-level regional numerical forecasts to the uncertainty in the specification of the initial state over the South American region. We perform several numerical experiments with the University of Utah Mesoscale Model in order to study the time evolution of the root mean square (rms) difference between the wind field predicted by a control forecast and a perturbed forecast. The initial state of the control forecast is the 700 Hpa geopotential field obtained by applying a regular multivariate objective analysis to the radiosonde data usually available over the region, and a 24-hr control forecast is performed. In order to obtain the initial state for the perturbed forecast, we degrade the original upper air data by an amount equal to the typical error measurement and perform the same objective analysis as before. We consider this way of introducing the uncertainty in the initial state as more realistic since the errors are introduced only in those points where the observations are available. Preliminary results show that the forecasts are sensible to the uncertainty in the specification of the initial state since the rms difference grows systematically with time. These findings are in contrast with previous studies that assign high predictability to low-level wind field predictions. In this context we perform air parcel trajectories analyses from different points in the domain and, in some cases, the differences are found to be significantly large. Although these are preliminary results that need further investigation, the sensitivity revealed may play a significant role in the South American region due to the sparsity of data.

3.5

REACTION KINETICS OF IODINE SPECIES IN THE TROPOSPHERE

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Methyl iodide appears to be the main source of iodine in the atmosphere and it has been observed in the marine boundary layer. We have used a discharge-flow resonance-fluorescence technique to study the rate and temperature dependence of the reaction between OH and CH_3I. Our results yield a rate constant of $(7.2 \pm 0.7) \times 10^{14}$ cm^3 $molecule^{-1}$ s^{-1} and an activation energy of (9.3 ± 1.7) kJ mol^{-1}. Thus we estimate that reaction of CH_3I with OH would account for 2% of the overall loss of methyl iodide, with the remaining 98% undergoing photolysis to form methyl radicals and iodine atoms. It appears that no other species containing iodine has been detected in the troposphere and sinks for the element are not known although HOI, HI and I on aerosols have been postulated.

We have observed a reaction between I_2 molecules and NO_3 radicals in the gas phase. The concentration of iodine was monitored using laser-induced fluorescence, and the concentration of nitrate radicals was measured using optical absorption at λ=662nm. Our data gave a second-order rate constant for the reaction of $(1.30 \pm 0.04) \times 10^{12}$ cm^3 $molecule^{-1}$ s^{-1}. It is interesting to note that there is no analogous reaction for the halogens F_2, Cl_2 and Br_2.

P2.1

LONG-TERM AEROSOL MEASUREMENTS AT THE SOUTH POLE

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The Climate Monitoring and Diagnostics Laboratory (CMDL) program of the National Oceanic and Atmospheric Administration (NOAA) conducts measurements of climatically important gases and aerosols at Amundsen-Scott South Pole Station. Condensation nuclei (CN) concentration has been measured since 1974, and aerosol scattering extinction (σ_{sp}) has been measured (using a four-wavelength nephelometer) since 1979. The CN data show an annual cycle with a maximum exceeding 100 cm^{-3} in the austral summer and a minimum of about 10 cm^{-3} in the winter. The σ_{sp} data show an annual cycle strikingly different from the CN record, being dominated by sea-salt events in the austral winter. Aerosol chemistry experiments were conducted during 1982 and 1987 that included aerosol filter samples for PIXE analysis, aerosol size distribution measurements in the Aitken size range, and aerosol black carbon measurements.

P5A.9

COVARIATION OF SOME COMPONENTS OF THE ANTARCTIC TROPOSPHERE

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Aerosol concentration varies with meteorological events on the south polar plateau. It has previously been shown that the bulk properties of the aerosol and the marine component of it are closelv related to temperature, wind speed and wind direction in the lower troposphere and bou iary layer. This paper discusses the concurrent variation of total aerosol, its marine and carbonaceous components, carbon dioxide, and meteorological parameters at the surface at the South Pole. Analyses of the response of concentrations to stagnation, storm circulation, decay of the circumpolar vortex, and the summer/winter transition are given.

P1.6

CHANGEABILITY OF SURFACE OZONE CONCENTRATIONS IN VARIOUS PHYSIOGRAPHIC AND WEATHER CONDITIONS IN THE ANTARCTIC

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During the "Trans-Ar . ctic Expedition" (27 July 1989 - 3 March 1990) the correlation between the surface ozone concentration and atmospheric temperature has been found. The correlation was absent when stratospheric cyclonic activity increased. In this case, stratospheric ozone intrusion was observed over the observational point. The dependence of ozone variations on the orography structure in the regions with complicated relief has been found as well.

The authors pay attention to the fact that the ozone measurements were carried out in the conditions of clean air. The data obtained can be considered as background ones.

5A.1

ASPECTS OF ANTARCTIC ATMOSPHERIC BEHAVIOR RELEVANT TO CHEMICAL COMPOSITION OF THE TROPOSPHERE

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The nearly perennial surface radiative temperature inversion over the sloping ice fields of Antarctica results in a continent-wide, radially diverging surface wind regime. The discharge of cold surface air across the coastline is concentrated into several zones about the continent. The divergence in the boundary layer is compensated by inflow aloft. This tropospheric and stratospheric convergence generates cyclonic rotation, and thus contributes to the development and centering of the circumpolar vortex over the highest parts of the continent. The time-averaged meridional mass circulation couples the surface layer directly to the stratosphere. The lighter surface winds around the time of the summer solstice imply a much weaker contribution to vortex forcing during summer than during winter.

Typically, synoptic-scale cyclones develop in midlatitudes, move poleward in a spiralling fashion, and reach the Antarctic coast in a dissipating state. The elevated ice sheet usually blocks the continued poleward motion of the storms. Upper level ridges, which disrupt the mean circulation, can direct the upper parts of coastal cyclones into the continent. Cases of entire storm systems moving over the relatively low-elevation ice sheet in West Antarctica have been documented. Recently, the combination of intensified surface winds from the ice sheet and synoptic forcing has been shown to lead to the very frequent formation of mesoscale cyclones just offshore; the climatic impact of these vortices is not yet known.

Precipitation in the coastal areas is episodic in conjunction with passing cyclones. Over the highest parts of the continent the semi-continuous fall of ice crystals from clear skies dominates the annual precipitation total. Orographically-induced lifting of moist air masses is the primary cause of precipitation formation over the ice sheet for all areas apart from the gently sloping high interior where radiative cooling predominates. Over East Antarctica, most precipitation falls during the winter, and little during the summer. A more complicated seasonal cycle is present over West Antarctica.

P4.1

AEROSOL AND GRAPHITIC CARBON CONTENT OF ANTARCTIC ICE CORE

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We have analyzed a set of 20 Antarctic ice core samples spreading in age over the last 20,000 years to determine the total amount of aerosols and black carbon (soot). Time variations provide information about the past tropospheric aerosol and soot loading, their changes in time and possible sources. Comparison with the available data of the present concentrations in fresh snow allows us to determine the current anthropogenic contribution to the total mass of atmospheric aerosols and black carbon.

For comparison, a similar analysis of the Greenland Camp Century ice core is presented. Similarity and differences between Antarctic and Greenland ice cores are discussed.

PI.17

VERTICAL PROFILES OF TRACE GASES IN THE ANTARCTIC TROPOSPHERE

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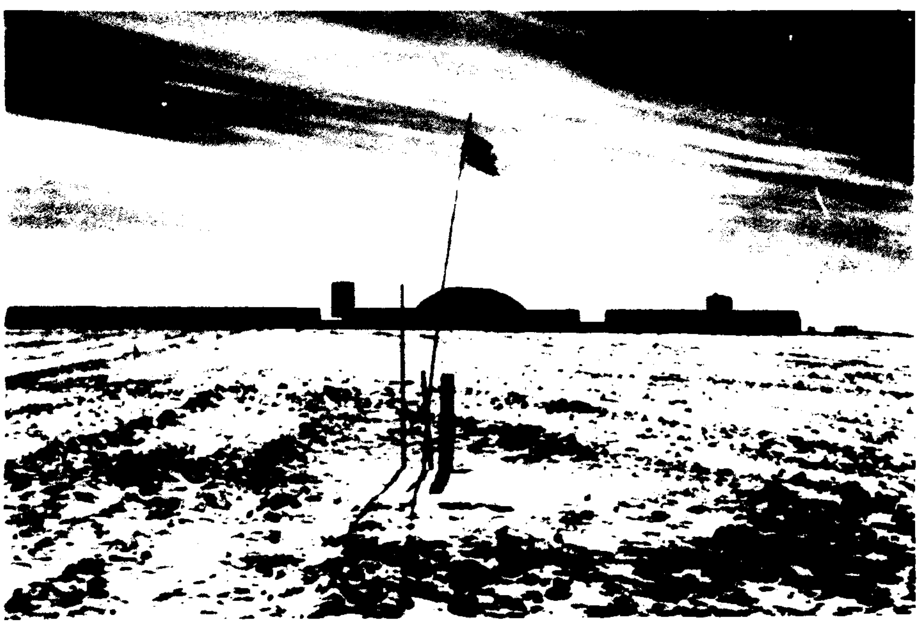
Air samples collected on helicopter flights to 15,000 feet and from ground level sites during three summer seasons near Scott Base (78°S) have been analyzed for a range of trace gases.

Stainless steel canisters were used and analysis was by GC/ECD and GC/FID in Australia and New Zealand. Successful collection and analysis was achieved for CH_4, CO_2, CFC-11, CFC-12, CH_3CCl_3, and N_2O, with sufficient results for at least preliminary indications of the amounts of CCl_4, CFC-113 and several NMHCs. The canisters proved unsuitable for the reliable storage of CO.

No strong vertical gradient was evident in the concentration data.

Comparisons have been made with concentrations of the trace gases as measured at Cape Grim (41°S) and Mawson (67°S) for the corresponding periods, and similar trends in trace gases are noted. In particular there is confirmation of the virtually uniform distributions of CH_4 and CO_2, the CFCs, and N_2O, through the Southern Hemisphere. The increasing trends of all gases between succeeding summers is virtually identical to those observed at Cape Grim.



New Pole Station under construction, 1974.

3.3

THE CHANGING COMPOSITION OF THE ANTARCTIC TROPOSPHERE: CARBON DIOXIDE AND METHANE DISTRIBUTIONS AND VARIATIONS

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The NOAA/CMDL cooperative global flask sampling network includes six mid- and high-latitude southern hemisphere sites: Amsterdam Island; Cape Grim, Tasmania; Palmer Station, Antarctica; Syowa Station, Antarctica; Halley Bay, Antarctica; and the Amundsen-Scott South Pole Station. Concentrations of the major greenhouse gases, CO_2 and CH_4 are measured in whole air samples collected approximately weekly at these locations. In-situ carbon dioxide measurements are also made continuously at the South Pole. Determining the global distributions of these gases is necessary in order to quantify the global carbon cycle and to understand how it will respond to global climate change.

The latitude distributions of annual means show that the lowest CO_2 and CH_4 concentrations on earth are found in the Antarctic. This is because the anthropogenic sources for both gases are predominantly in the northern hemisphere. However, because of their relatively long lifetimes, the concentrations of both gases are increasing in the Antarctic at rates similar to those in the northern hemisphere: ~1.4 ppm CO_2 yr^{-1} and ~12 ppb CH_4 yr^{-1}.

A detailed analysis reveals that CO_2 concentrations in the northern hemisphere increased faster during 1988 and 1989 than in the southern hemisphere, leading to a significant change in the interhemispheric gradient compared to 1981-87. Is this a temporary anomaly or the first indication of a change in the global carbon cycle? Closer scrutiny of the CH_4 data suggests that a decrease in the CH_4 growth rate has occurred during the period 1983 to 1989. Does this reflect a change in CH_4 emissions or in the photochemical CH_4 sink?

The answers to these questions are crucial to our ability to predict and understand the effects of atmospheric and climate change. The Antarctic troposphere CO_2 and CH_4 measurements will play a key role in unravelling the complex atmosphere-ocean-biosphere processes comprising the global carbon cycle.

P5A.7

TROPOSPHERIC CLOUD STATISTIC FROM LIDAR DATA IN DUMONT D'URVILLE (66°S, 140°E) FOR THE YEAR 1989

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A backscattering (and depolarization) lidar operated at 532 nm over the years 1989 and 1990 in the French base of Dumont d'Urville (Antarctica). The instrument carried out both continuous (routine) measurements and intensive measurements (during the periods of ECLIPS campaigns in April and September-October) of tropospheric and stratospheric clouds. ECLIPS (Experimental Cloud Lidar Pilot Study) is an international campaign of ground based lidar and satellite cloud measurement devoted to obtaining cloud parameters useful for climate modellers.

Lidar data were supported by daily rawinsonde soundings from the same site, by continuous ground-based meteorological observations (e.g. cloud coverage, cloud type, etc.), and by videotape recording of the sky.

The elaboration of almost one year (1989) of such data (the lidar instrument did not operate with adverse weather) gave an archive and a statistic of cloud optical properties at 532 nm (optical backscattering and extinction coefficients, optical thickness, depolarization) and of macrophysical characteristics (cloud base, cloud top, midcloud and relative meteorological parameters like temperature, relative humidity with respect to water and ice, wind speed, direction and shear, advection, etc.).

A plot of 1989's meteorological data trend at Dumont d'Urville (derived from rawinsonde soundings) was produced, too. A review of results obtained will be shown, with special regard to macrophysical properties of tropospheric clouds. Some particular cloud events will be discussed.

Aerosol observation upwind of Old Pole Station, 1 February 1974.

4.4

GLACIOLOGICAL STUDY OF THE ANTARCTIC ATMOSPHERE PERTURBATIONS CAUSED BY VOLCANIC ERUPTIONS

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Cataclysmic volcanic eruptions disturb markedly the global atmosphere for generally two years. Long-life volcanic products (mainly H_2SO_4), stored in the stratosphere (Junge layer), come back slowly to the troposphere. Antarctic snow is then contaminated and acid signals are recorded. The recovery of these signals along Antarctic ice cores provides a history of past volcanic events, most generally of global, but sometimes of just regional, significance.

Our presentation is mainly based on the analysis by several physical or chemical techniques of a 1000-year ice core drilled near Amundsen-Scott Base. Acid and ultrafine ash deposits of volcanic origin have been carefully investigated. Major volcanic eruptions were detected, dated and tentatively identified. Comparisons with similar Antarctic and Greenland records have been frequently used. It was found that the imp ct of major volcanic eruptions is only significant but very sporadic on the global atmospheric sulfur budget. The chlorine cycle and the hemispheric climate, as recorded in Antarctic ice, seem to be little affected even by such a large event like Tambora (1815).

The amount of volcanic sulfate deposited in Antarctic snow, averaged over the last millenium, is, however, minor (13% of the total sulfate) in comparison with that of the marine biogenic source. The study suggests also that the 19th century was the period of the millenium most seriously disturbed by global explosive volcanic activity.

6.5

TRANSPORT AND DEPOSITION PROCESSES OF AEROSOL PARTICULATES IN THE ANTARCTIC TROPOSPHERE

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The ability to interpret the changes in chemical composition along ice cores in terms of changes in atmospheric composition is currently limited by a rather poor knowledge of air-to-snow transfer processes. The variety of processes involved and the influence of weather and climate on these, have made the study of the air/snow relationship very difficult. This is particularly so in the very remote and clean Antarctic environment.

This paper reviews our present knowledge of air-to-snow transfer processes for aerosol particulates in clean air locations. The problems of understanding the sources, reactions, transport and deposition of aerosol particulates are discussed and gaps in our current knowledge are highlighted. Experimental methods by which some of these problems might be solved are suggested, and the difficulties involved in applying such methods in Antarctica are discussed.

P6.5

WET DEPOSITION OF AEROSOL COMPONENTS OVER THE ROSS ICE SHELF

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Aerosol and fresh surface snow samples have been collected at a remote site on the Ross Ice Shelf, Antarctica. Parallel analysis of these samples for sea-salt, crustal dust, sulfate and nitrate components has been carried out, allowing scavenging ratios to be estimated. Results are compared with data from the Antarctic Peninsula, taking into consideration the climatic differences between the two regions.

P5B.3

ON THE ESTIMATION OF SOME EFFECTS OF AEROSOLS ON CHEMISTRY AND THE UV-VISIBLE RADIATION FIELD OF THE ANTARCTIC TROPOSPHERE

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Tropospheric aerosols can be responsible for heterogeneous ozone depletion due to the downward motion of Antarctic air masses. Possible schemes of such heterogeneous processes are discussed and evaluated using a 1-D photochemical model. The impact of polar stratospheric clouds (PSCs) on tropospheric UV-visible radiation and chemistry is studied. The radiation and chemical effects at twilight are of special interest. The twilight UV-visible radiation fields obtained by our version of the delta-Eddington two-stream scheme are shown to be in good agreement with the more accurate spherical code results (Anderson and Lloyd, 1990). For solar zenith angles greater than 90 degrees the effects of trapping the twilight solar radiation below the cloud are estimated.

P1.4

STUDY OF ANTHROPOGENIC IMPACTS ON TROPOSPHERIC TRACE GASES IN THE ANTARCTIC

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The impact of anthropogenic disturbances on composition and temperature of the Antarctic troposphere is investigated with a two-dimensional radiative photochemistry model.

It is shown that the largest changes in chemical composition in the Antarctic resulting from discharges of CO, CH_4, N_2O, and chlorofluorocarbons such as CF_2Cl_2, $CFCl_3$ and CCl_4 take place at the pole. The variations observed are maximal for ozone: its significant increase at heights ~4-10 km has been determined. As a consequence, temperature in this range essentially increases, too.

With atmospheric discharges of methane, water vapor content increases considerably in Antarctica, especially in the middle and lower stratosphere. Such a manifestation of CH_4 discharges leads, first, to a greenhouse effect two times that of just enhanced CH_4 content, and, second, to greater polar stratospheric cloud occurrence in the cold Antarctic lower stratosphere in winter.

[P5A.1]

THE SOUTH POLAR PLATEAU — THE ULTIMATE SITE FOR TROPOSPHERIC MONITORING

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A cold dry air mass is permanently resident over the South Polar Plateau. This air mass is uniquely derived from cooling and drying of mP air which encircles the globe about the periphery of Antarctica. The uniqueness of this air mass is evidenced by the similarity in potential temperature at the many stations in the air mass, and the very small annual variation of CO_2 and H_2O concentrations observed on the Plateau. This paper discusses meteorological and atmospheric chemical processes over and around Antarctica, which provide this unique setting for monitoring tropospheric composition.

Recent and historic experiments and observations indicate interaction among CO_2 and H_2O in the air. For example, Hogan et al. (1991) have presented data that show the smallest observed amplitude in seasonal variation in CO_2 and H_2O vapor concentrations occur at the South Pole. Data are obtained from monitoring stations for which systematic measurements are available. The reason for this small co-variation was pointed out by Egan et al. (1990) in that there is a linkage between CO_2 and H_2O vapor. Thus, where there is a small seasonal variation in H_2O vapor, the seasonal variation in CO_2 concentration will be small. Water vapor concentration above the Polar Plateau is confined to a narrow range because the air temperature rarely exceeds -25°C. During the spring, displacement of the cA air mass over the South Pole by an mP air mass occurring at the time of major temperature differences in these air masses results in air with a dewpoint of -35°C or less being replaced by air with a dewpoint of -20°C or more. The resulting H_2O vapor pressure difference is only about 1 mb, with a resulting annual range of CO_2 concentration of ~2 ppm.

In addition, diffusion coefficient measurements of H_2O vapor in air and CO_2 made in European laboratories in the late 1800s show a CO_2–H_2O vapor linkage. This linkage is confirmed in laboratory high resolution Fourier transform spectroscopic experiments performed at City College/CUNY using CO_2 and H_2O vapors.

These and other interactions will be discussed, to illustrate possible inferences of global tropospheric composition from South Polar observations.

3.1

MEASUREMENTS OF NITROUS OXIDE AND CHLOROFLUOROCARBONS BY IN-SITU GAS CHROMATOGRAPHY AND FROM FLASK SAMPLES COLLECTED AT SOUTH POLE

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Atmospheric nitrous oxide (N_2O) and the chlorofluorocarbons (CFC's) are receiving considerable attention because of their role in greenhouse warming and ozone depletion. Measurements of these gases at South Pole (SPO) are critical to our understanding of global climate change, because this location is one of the few places in the world that is completely removed from local sources. We have measured nitrous oxide, CFC-12, and CFC-11 from flask samples collected at SPO since 1977. Although flask samples provide an adequate record of long-term growth rates, finer time resolution and better precision can be achieved with in-situ gas chromatographs (GC's) to resolve issues involving stratospheric ozone depletion and atmospheric circulation. It has been suggested that vertical circulation above Antarctica may influence the lifetimes of atmospheric trace gases. In 1988, we added an automated GC system at SPO to measure N_2O, CFC-12, CFC-11, methyl chloroform (CH_3CCl_3), and carbon tetrachloride (CCl_4) concentrations in the atmosphere once every two hours. We are now detecting subtle variations in halocarbons (<2 ppt for CFC-11) which may help resolve these issues. We will report long-term growth rates and compare our data with other atmospheric parameters to identify sources of these gases into the Antarctic region.

4.6

CHANGES IN TROPOSPHERIC METHANE BETWEEN 1841 AND 1978 FROM A HIGH ACCUMULATION-RATE ICE CORE

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Methane is an important component of the background troposphere because of its roles in the chemistry of ozone, hydroxyl radicals and carbon monoxide. It is also a greenhouse gas that has doubled in concentration since pre-industrial time (about 1800 AD) causing an increased radiative forcing of 28% relative to that of carbon dioxide. Global sampling networks have characterized the temporal and spatial distributions of methane since 1978 and air occluded in polar ice has normally recorded tropospheric methane concentrations from glacial times until the middle of this century. Results reported here are from a new ice core containing air with a remarkably fine age resolution that fills the gap between these two types of records, where previously only sporadic and inferred data existed. The ice core (DE08) was drilled from a region of Law Dome, Antarctica, where the large snow accumulation rate of 1200 kg m^{-2} yr^{-1} quickly occludes air into bubbles. The air is generally 35 years younger than the ice and the air-age distribution width is less than 8 years. Melt features in the core are rare and did not noticeably affect the air enclosure process or chemistry. A dry air-extraction technique and gas chromatography were used to find the methane concentrations from 1841 to 1978 every 10 years or less with a precision of ±20 ppbv (parts per billion volume). The rate of increase of methane concentration during the period 1965-1975 was probably greater than observed since 1978 when the modern direct record from Cape Grim (Tasmania) began. The overlap of the DE08 ice core record with the Cape Grim record implies that there is no significant modification to methane during the enclosure and storage of air in the ice or during its recovery and analysis.

P6.4

DIRECTLY OBSERVED HEAT AND MOISTURE EXCHANGES AT SOUTH POLE STATION

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The turbulent fluxes of heat, moisture, momentum and aerosols to and from the surface are important components of larger-scale budgets. Episodes of aerosol deposition, for example, must certainly be coincident with events of turbulent momentum and heat flux. The turbulent fluxes appear as small but important terms in a heat balance that consists of large and largely cancelling radiation terms. That is, though incident and reflected short-wave radiation and downwelling and upwelling long-wave radiation are each large in magnitude, their sum is quite small. Thus, traditional methods of inferring turbulent heat and moisture fluxes by apportioning the net radiation may be subject to large errors, and alternate methods are preferable.

Direct eddy-correlation observation of the turbulent fluxes at remote sites is now technically feasible, and perhaps even more convenient than traditional gradient methods. We operated a sonic anemometer, a fine-wire thermocouple, a fast-response hygrometer, lightweight horizontal and vertical propeller anemometers at South Pole Station during 1988. In addition, a digital whole-sky image was obtained every five minutes during January. Direct eddy fluxes were obtained during January and February. After that time, fluxes were estimated using the propeller anemometers and empirical flux-variance relations obtained during the earlier period. Inspection of power spectra of wind and temperature signals verifies that surface layer turbulent motions were observed. Although some uncertainty remains about how well the hygrometer functioned, temporal consistency between moisture fluxes and the momentum and heat fluxes indicates that we obtained direct measurements of the latent heat flux on the Antarctic plateau. Our analysis included study of the boundary layer thickness and strength based on radiosonde launches made at Pole Station.

Conclusions based on our observations include:

1) Moisture fluxes and heat fluxes correlate strongly with wind direction, itself an indicator of air mass type arriving at the station. In January, long periods of free convection are observed.

2) Extensive time series reveal that H and LE have magnitudes that rarely exceed 10 W/m^2, and that episodes of appreciable flux last up to 15 hours. In most cases, the observed turbulent fluxes are small enough that it would be hard to obtain reasonable estimates using Bowen ratio methods. The distributions of hourly values of H and LE during January are positively skewed, with the median value of H at 1 W/m^2 (with extremes at -6 and +12 W/m^2) and LE at -2 W/m^2 (with extremes at -10 and +20).

3) Moisture flux from the surface associated with changing air mass type is observed up to four hours prior to fog forming at Pole Station.

P2.13

MODELING THE STABLE ISOTOPIC COMPOSITION OF PRECIPITATION

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A two-dimensional advection model with parameterized cloud and isotopic microphysics has been developed and applied to typical cross-sections observed in extratropical cyclones. The model is designed to simulate and explain the isotopic composition of precipitation. The model uses imposed velocity and temperature fields. Six species of water and their various interactions are included in the microphysical routine. The isotope equations include the effects of evaporation, equilibration and nonequilibrium condensation.

5A.2

DESERT DUST AND SEA-SALT AEROSOL IN ANTARCTICA: SIMULATIONS WITH A GENERAL CIRCULATION MODEL

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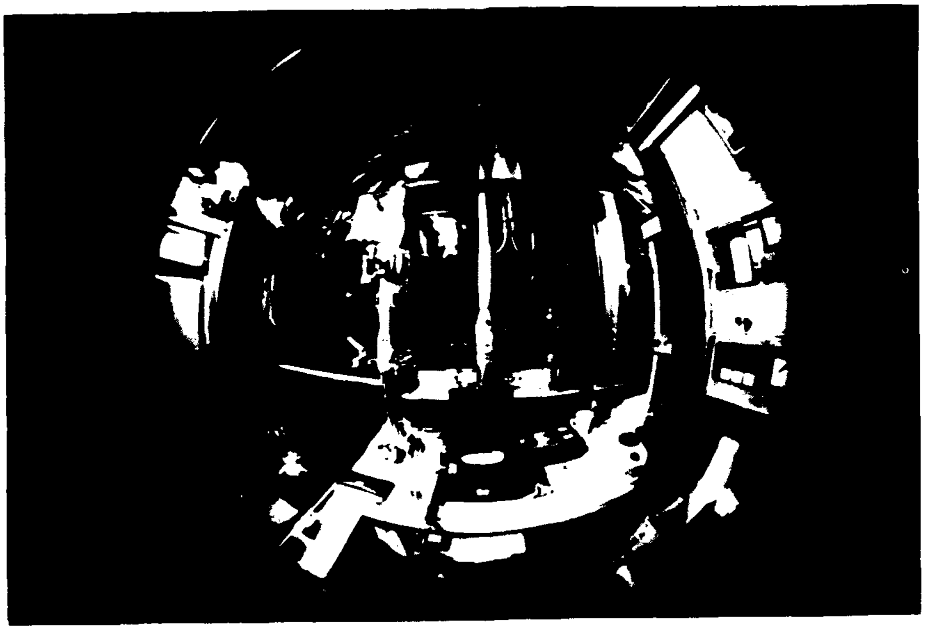
Parameterizations for the sources, mobilization, transport and deposition of desert dust and sea-salt aerosols in the atmosphere have been developed within the Goddard Institute for Space Studies (GISS) General Circulation Model (GCM). The strong observed seasonal cycle of both species in Antarctica is well reproduced by this model. Ice cores have revealed large variations in polar regions on time scales of glacial-interglacial cycles as well. Possible causes for such variations are multiple, e.g.: changes in the sources location and extent, in the source production efficiency, in the intensity of the atmospheric transport, etc. Coupled aerosol/climate ice age simulations help analyze and weight these contributions. They therefore provide a unique insight on the meaning of some of the most striking ice core records of climate and environment changes.



Komhyr sampling stack, Pollak counter, ozone meter, recording system in aurora hut, Old Pole Station, 1974.

P1.2

OZONE CONCENTRATION AND MICROMETEOROLOGICAL MEASUREMENTS IN THE ATMOSPHERIC SURFACE LAYER AT "CAMPO ICARO" BASE IN ANTARCTICA

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During the Italian Scientific Expeditions in Antarctica in 1987-88 and 1988-89, some measurements of micrometeorological parameters and ozone concentration were performed at "Campo Icaro" close to the Italian station of Terra Nova Bay (74.44°S, 164.05°E) in order to determine some characteristics of the atmospheric surface layer and to estimate ozone deposition rates at the site[1].

In areas of the Antarctic region near the coast, such as the Italian base, the presence of bare soil increases net radiation values. This modifies the thermal structure of the atmosphere at the lower layer, causing convective motions.

The energy balance on clear summer days leads to a local circulation characterized by convective cells. This small-scale convective motion basically involves an alternation between ascending and descending flows. Temperature measurements at the ground up to 10-m height show a highly pronounced superadiabatic profile that can justify this behavior pattern. These convective cells have been measured along the coast during summer months by vertical wind intensity sensors installed on a pole[2], and by SODAR measurements[3].

The ozone concentration measurements performed, using a UV absorption photometer DASIBI, in the first 10 meters of the atmosphere, showed a maximum when the sun reaches its zenith in the diurnal variation trend that could be explained with photochemical processes in the lower layer. These processes are remarkably intensive, in fact, the difference between maximum and minimum is of the same magnitude of the diurnal average concentration value. It would be of great interest to perform contemporary measurements of nitrogen oxides and nonmethanic hydrocarbons in order to assess the role of photo-oxidation processes in the formation of the Antarctic tropospheric ozone.

In unpolluted areas, such as high mountains, the ozone concentration values due to photochemical formation does not reach as remarkable an amount as that measured at the Italian base of Terra Nova Bay. Furthermore, a diurnal variation of such a large extent needs additional investigations in order to understand if the causes of production are due to natural or anthropic effects. Natural causes must be investigated in all phases including large scale vertical transport phenomena and photolytic phenomena, the latter because of the extreme atmospheric transparency. Anthropic effects could be due to transport of primary pollutants on a local or global scale; this transport could supply chemicals for photo-oxidation reactions enhanced by the great amount of solar radiation that reaches the atmospheric surface layer.

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2. Georgiadis, T., P. Bonasoni, and G. Giovanelli, 1990. Characterization of the Antarctic surface layer at the "Campo Icaro" base in Antarctica: A case study, 2nd Workshop Italian Research on Antarctica Atmosphere, *SIF Conf. Proc., 27*, 35-44.

3. Mastrantonio, G., R. Ocone, A. Pellegrini, and G. Fiocco, 1990. Sodar observations of the Antarctic boundary layer in a deglaced area: Preliminary results, *Il Nuovo Cimento, 13c*, 589-597.

6.3

CHEMICAL FRACTIONATION OF SEA SALT IN THE ANTARCTIC ATMOSPHERE

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The SO_4^{2-}/Na^+, SO_4^{2-}/Cl^-, SO_4^{2-}/Mg^{2+} ratios in deposited snow in the coastal area of Antarctica, west coast of Norway, and on Spitzbergen are markedly lower than for sea water. (Deficit of sulfate with regard to sea water.) Precipitation and newly fallen snow in summer in the same area have an excess of sulfate relative to sea water. More than 50-km inland from the coast there is an excess of sulfate. Near the coast the deficit of sulfate relative to sodium is of the order of 300 mg m^{-2} yr^{-1}.

The explanation of this phenomenon is as follows: Droplets of sea water in the atmosphere formed by breaking waves in strong wind have residence time in the atmosphere of minutes to hours. At low temperatures these drops may freeze rather than evaporate. During the freezing there will be a small pocket of brine in the ice crystal, and the ion concentration in this brine will increase as the temperature decreases. Laboratory experiments with sea water have shown (Richardson, 1976) that Na_2SO_4 $10H_2O$ starts to precipitate at about -9°C, $CaSO_4$ $2H_2O$ at -10°C, and NaCl $2H_2O$ at -22°C. The SO_4^{2-}/Na^+ ratios in the brine at -10°C and -20°C were, respectively, 0.146 and 0.021. Some of this brine will drain from the frozen sea droplet and be transported inland while the frozen sea droplet with a mass some 100 times the brine will be deposited in the sea. This chemical fractionation process in the atmosphere takes place at low temperatures and leads to brine droplets in the atmosphere with a deficit of SO_4^{2-} relative to the other main ions. Chemical analysis of aerosols in the Arctic in winter confirms this.

USARP quarters, Old Pole Station, 1974.

2.1

ATMOSPHERIC NUCLEI IN THE ANTARCTIC TROPOSPHERE: THEIR DISTRIBUTION, NATURE AND ORIGIN

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Systematic series of observations of surface-level condensation nucleus concentration have been made at several sites on the Antarctic continent. The longest series, which was started in 1974, is from the USA Amundsen-Scott Station at the South Pole. Coastal observations have been made from the Australian Station Mawson since 1981 and the German Station Georg von Neumayer since 1982. Shorter series of measurements have been reported for other sites, in particular from the Japanese coastal Station Syowa from 1977 to 1978.

The very pronounced and characteristic seasonal cycle in concentration observed over Antarctica and at other remote southern locations has been used to infer the dominant role of photochemistry in the production of these remote atmospheric nuclei. More recently systematic measurements of aerosol chemistry from these long-duration measurement sites, confirm the dominant role played by sulfate particles of marine origin during the active particle production period (austral summer).

Some information on the seasonal variation of nucleus size distribution has been reported previously for Syowa and data are now available from several years of diffusion battery measurements at Mawson. There is a non-linear relationship between non-sea salt sulfate and nucleus concentration. The combined nucleus concentration, size distribution and chemistry measurements, however, indicate a consistent picture of evolution of the size distribution and growth, and decay, of the submicrometer aerosol mass.

Interannual variation in nucleus concentration at the coastal station Mawson is a significant component in the total variability and complicates the determination of any long-term trend. There is some evidence that suggests that large scale climate features may be influencing nucleus concentration, although whether this is through production, transport or loss is still an open question.

PI.7

SOME RECENT RESULTS OF SOVIET MEASUREMENTS OF SURFACE OZONE IN ANTARCTICA: A METEOROLOGICAL INTERPRETATION

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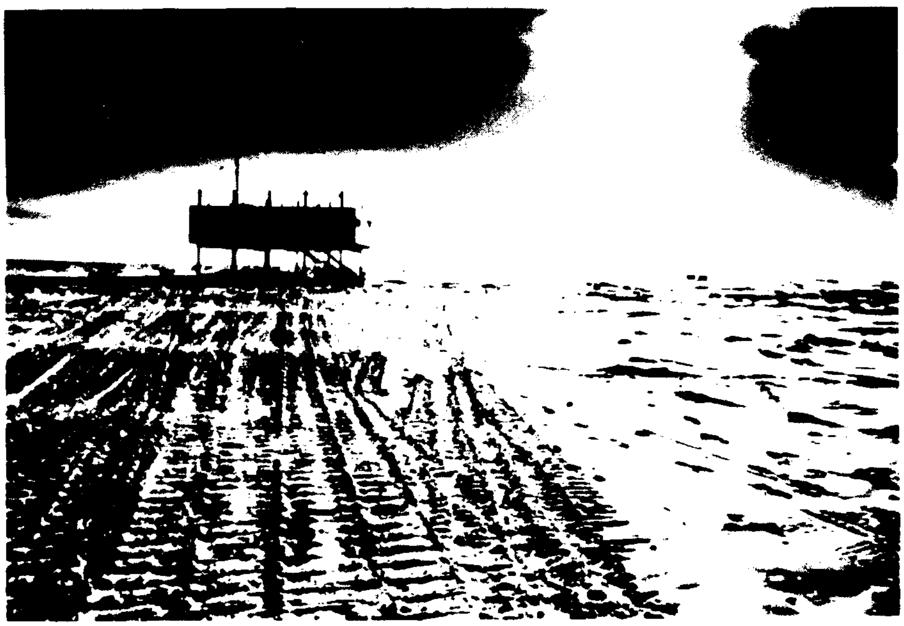
Surface ozone measurements were made at Molodezhnaya (67.40°S, 45.50°E) and Mirny (66.33°S, 93.01°E) stations in spring 1987 - autumn 1988 with the Dasibi AH-1008 ozone analyzer. The data show an annual variation with a summer minimum of 15 ppbv value. The daily-mean surface ozone content has lowered on some summer days down to 9 ppbv. The striking feature of the surface ozone record is two types of day-to-day variability. One of the types is characterized by large day-to-day variations with a range rising up to about 10 ppbv. The probable mechanism of such variations is the vertical transport induced by cyclone activity. So the surface ozone correlates strongly with the temperature (positively) and the relative humidity (negatively) in the troposphere above the boundary layer. In the synoptically quiet period, late in summer, the day-to-day ozone variations were not so large. The most likely mechanism of these variations is the slope catabatic wind which transports ozone from inside the Antarctic continent. The ozone content usually increased when the surface wind had turned to the southern direction. The near-surface ozone was also measured aboard an airplane along the path from Mirny through Vostok (78.28°S, 106.48°E) to Mirny in a synoptically quiet period. The plane flew at the altitude of a few tens of meters above the Antarctic plateau. The ozone content increased slightly toward Vostok. This confirms the hypothesis of slope wind dependence of surface ozone. The surface-ozone record was also analyzed by the spectral method and the method of fractal analysis. The correlation dimension of dynamical attractors was calculated for different types of ozone variability.



Clean air facility, first days of measurements, summer 1976–77.

P5B.1

TROPOSPHERIC OZONE ANNUAL VARIATION AND POSSIBLE TROPSPHERE-STRATOSPHERE COUPLING IN THE ARCTIC AND ANTARCTIC AS DERIVED FROM OZONE SOUNDINGS AT RESOLUTE AND AMUNDSEN-SCOTT STATIONS

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A tropospheric ozone annual variation in the northern and southern polar regions is analyzed on the basis of the data of ozone soundings at Resolute (74.41°N, 94.59°W) station for 14 years (1974-1987) and Amundsen-Scott (South Pole) station for 7 years (1967-1971, 1986-1987). The ozone annual variation in the free troposphere above Resolute is connected with that in the lower stratosphere. From the middle troposphere up to the lower stratosphere the changes of ozone concentration and ozone mixing ratio vertical gradient have the same tendency, the latter having its maximum variation at the tropopause level. At the same level the maximum of the wind vertical shear and the maximum of the static stability variation are noted. During April-May there is the most significant increase in the middle and lower tropospheric ozone mixing ratio, which is probably connected with the stratospheric injections. The ozone annual variations in the surface and boundary layers differ strongly from that in the overlying troposphere. This is most likely due to photochemical processes which proceed more intensively under the lower tropospheric temperature inversion.

The relationship between tropospheric and lower stratospheric ozone above the South Pole changes during the year. So the ozone annual variations in the troposphere and lower stratosphere are different in winter. In spring, during the development and filling up of the ozone hole, they are similar, which points to the stratosphere-troposphere coupling in the season. The ozone concentration and mixing ratio vertical gradient in the middle troposphere change in a similar manner during the whole year, but in the upper troposphere their variations are contrary (except the time during the ozone hole). The change of the sign of this relationship occurs approximately at 6-7 km altitude. At the same level the maximum variations of the ozone mixing ratio vertical gradient as well as the maximum variations of the static stability and the maximum of the wind vertical shear are noted. The significant feature of Antarctic ozone is also the contrary ozone annual variations in the lower and upper troposphere.

P2.2

GEOCHEMICAL CHARACTERIZATION OF AIRBORNE PARTICLES FROM THE LOWER TROPOSPHERE OF TERRA NOVA BAY, ANTARCTICA

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Major and trace elements (soluble + insoluble) of wind-blown particles collected at Terra Nova Bay were studied and the data compared with lacustrine and marine sediments of the same area and the Ross Sea.

Estimated dust loadings range from 0.06 to 0.91 μg/m^3, and the insoluble fraction represents 5 to 43% of the total. The TSP concentration shows an increase with winds turning from N-NW to E-SE, and variable values are mainly related to calms and mixed barrier/marine winds.

The Transantarctic Mountains (granitoids and metamorphic rocks) are the main sources of the large (>4μm) particles, with a possible contribution from nearby volcanoes.

Some evidence of anthropogenic contamination was found in the samples, with mean Pb enrichment factor (EF_{crust}) value of 63 (concentration range 19-62 pg/m^3).

The comparison with coastal and offshore marine sediments shows some similarities of the aerosol particles with offshore samples collected near Coulman Island and no evident relation with the sediments of four small lakes collected in the area.

Aurora hut and balloon inflation shelter, as seen from the head of the Holy Stairs. Old Pole Station, January 1974.

2.7

AEROSOL BLACK CARBON MEASUREMENTS AT SOUTH POLE OBSERVATORY: IDENTIFICATION OF LONG-RANGE AEROSOL TRANSPORT EVENTS

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Aerosol black carbon ("BC") is a long-lived tracer species distributed throughout the troposphere. It has no known noncombustion sources, either natural or anthropogenic. Its presence at the South Pole is an indicator of long-range aerosol transport, as the nearest plausible source regions are located on the continents of the Southern Hemisphere. Continuous measurements of BC have been made at SPO since December 1986. Data is recorded on a one-hour timebase, although it is necessary to average over 12 to 24 hours in austral winter when concentrations are very low. We find that the data record contains periods of distinct excursions in the BC concentration. During these events, the BC level rises above its adjacent "background" level by a statistically significant amount, over a period from a few hours to a few days. Some of these events are gigantic, and are clearly due to contamination from local sources at SPO. Other of the events occur at times when there appear to be incursions of distinctly different air masses. These meteorological features may be injecting "polluted" air from lower latitudes into the polar plateau.

We discuss the analysis of the data as an indicator of these transport events. These events may then be correlated with excursions in other measured atmospheric properties and with trajectory analyses, to assist in the identification of the means whereby lower-latitude air masses are transported across the pole.

The presence of optically-absorbing black carbon in the Antarctic troposphere implies the possibility of its deposition to the snow surface, lowering the surface albedo and changing the solar radiation balance. Thus, BC may have an effect in its own right, in addition to serving as a tracer for long-range aerosol transport.

Finally, we shall pose (but not answer) the question: What is the transport pathway for the lowest-ever concentrations of BC measured in the depths of austral winter?

Portions of this work were supported by the US Dept. of Commerce, NOAA.

6.4

DEPOSITION OF SULFATE AEROSOL TO THE ANTARCTIC SNOW SURFACE

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Most cloud condensation nuclei in the troposphere are sulfate particles. They have been postulated to be a significant factor in global temperature control through the cooling effects of backscattering and cloud albedo. This hypothesis is supported by ice core analysis which shows large variation of $SO_4^=$ and a negative correlation between isotopically inferred temperature and $SO_4^=$ concentrations in the ice. Sulfate appears to reach maxima during ice ages and minima during interglacial periods. The $SO_4^=$ concentrations in ice are believed to reflect historical tropospheric $SO_4^=$ concentrations.

Samples of air, falling snow, and fallen snow will be collected at the South Pole during the Austral summer of 1990-91 and analyzed for $SO_4^=$ using ion exchange chromatography in order to investigate the relationship of current snow $SO_4^=$ concentrations, and current tropospheric $SO_4^=$ concentrations and the efficiency of "wet" deposition. Condensation nuclei concentrations will be measured at the snow surface and in the interstitial air within the snow to investigate the importance of dry deposition of $SO_4^=$ into the snow.

5A.4

SOUTH POLE CLIMATOLOGY

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Since 1974, CMDL/GMCC has made hourly-average observations of meteorological variables (surface wind, station pressure, temperature, and humidity) at the South Pole Observatory (SPO) to assess the potential for local contamination of aerosol and atmospheric chemistry measurements and to reference these measurements to standard atmospheric conditions. Variations in the monthly and annual means of these observations will be presented as representing the climatology of the SPO. Where possible, the CMDL/GMCC results for the previous 15 years will be compared to the extended station record of 33 years. The climatology of the occurrence of "clean air" sector winds will also be presented as an indicator of the bias introduced in data by using such editing criteria. Since temperature gradient measurements were begun in 1984, lapse conditions have occurred on many occasions and for extended periods, even during the polar night. A pronounced wind shift often accompanies such episodes. When considered together, often in the form of the bulk Richardson number, the surface wind and temperature gradient represent a way of evaluating the degree to which the surface layer flow is coupled to the large-scale flow aloft. The seasonal and inter-annual variations of these occurrences will be presented. And in the few instances for which there are valid moisture observations, they will be presented as well.

P5A.9

COVARIATION OF SOME COMPONENTS OF THE ANTARCTIC TROPOSPHERE

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Aerosol concentration varies with meteorological events on the south polar plateau. It has previously been shown that the bulk properties of the aerosol and the marine component of it are closely related to temperature, wind speed and wind direction in the lower troposphere and boundary layer. This paper discusses the concurrent variation of total aerosol, its marine and carbonaceous components, carbon dioxide, and meteorological parameters at the surface at the South Pole. Analyses of the response of concentrations to stagnation, storm circulation, decay of the circumpolar vortex, and the summer/winter transition are given.

6.6

PARTICLE EXCHANGE TO THE ANTARCTIC SNOW SURFACE: THE BEGINNING OF A TROPOSPHERIC HISTORY

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The sodium concentration measured in recent South Polar snow by LeGrand and Kirchener (1988) has been compared with surface aerosol measurements (Bodhaine and Shanahan, 1990) made at the South Pole Station. When a 180-day lag is applied to allow spring/summer aerosol to be precipitated and incorporated into the autumn/winter snow layer, very good correspondence exists among mean aerosol concentration in air and mean sodium concentration in snow. This paper described analyses of meteorological and glaciological processes necessary to the exchange of particles from air to snow.

The accumulation rate of snow on the South Polar Plateau has been established through pit and core analysis at several times during the occupation of the station, and through direct snow stake accumulation measurement. Precipitation has not been routinely measured, but is estimated through analysis of the station meteorological record and application of the cold air precipitation rates derived from literature. The estimated precipitation is compared with measured accumulation to investigate lag between time of precipitation and accumulation and to examine other possible sources of accumulated ice substances (riming drifting and direct sublimation).

Precipitation of sodium, and other particles, through coagulation and other ice crystal attachment processes is discussed. The results of this work indicate that additional ground truth relative to the gradients of temperature, wind, precipitation and accumulation on the polar plateau are essential to the formal establishment of a theory relating glaciochemical accumulation to meteorological processes.

2.2

THE SIZE DISTRIBUTION OF SUBMICRON AEROSOL IN THE ANTARCTIC TROPOSPHERE

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The average size (January 1977 to January 1979) and size distribution (August to December 1978) of Aitken nuclei was observed with the automated Pollak counters and diffusion batteries at Syowa station, Antarctica. Daily mean size distribution was mostly bimodal having a trough at 0.01 μm in radii. The comparison with the seasonal variations of average size suggests that the height of the larger mode of the size distribution is maximum in summer and minimum in winter whereas the smaller mode is mostly constant during spring, summer and autumn and it disappears in winter.

In the separate program, the monthly average size distribution of sulfuric acid particles was evaluated by use of an electronmicroscopic technique for the samples obtained at Aska Station, Antarctica from January to May in 1988. Sulfuric acid particles were detected in the size range of larger mode of Aitken nuclei showing the seasonal variation similar to one suggested above. The size range of smaller mode of Aitken nuclei was out of detection limit of microscopic technique.

The smaller mode would be maintained by new particle production at a rate of about 100 particles per cubic meter per second. Assuming binary vapor system of sulfuric acid and water, such production rate would be attained by the photochemical production of sulfuric acid vapor from sulfur dioxide of 5 μg per cubic meter in the oxidation rate of 1% per hour.

P5B.5

NIGHT-TIME OXIDATION OF DMS: PRODUCTS, MECHANISM AND GAS-PARTICLE INTERACTIONS

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The night-time oxidation reaction between dimethylsulfide (DMS) and the nitrate radical (NO_3) in air has been investigated using two different experimental set-ups.

The products and mechanism were studied in a 480-L teflon-coated glass cell equipped with an on-line FTIR spectrometer and with ion chromatography off-line. The main end products were found to be methanesulfonic acid, SO_2, H_2SO_4, HNO_3, CH_2O, CH_3ONO_2 and CH_3SNO_2. The mechanistic study indicated that the three radicals ($CH_3S\bullet$, $CH_3S(O)\bullet$ and $CH_3S(O_2)\bullet$) are key intermediates in the oxidation of DMS in the troposphere.

The gas-particle interactions were studied in a 1300 L teflon bag with an O_3-monitor, a SO_2-monitor, a DMPS (differential mobility particle sizer) and a CNC (condensation nuclei counter) on-line and both the gas phase and the particles were analyzed by ion chromatography off-line. We found that methanesulfonic acid is not as liable to nucleate as H_2SO_4 and that the accommodation coefficient α_{MSA} for methanesulfonic acid to stick on H_2SO_4 particles is much lower than $\alpha_{H_2SO_4}$ for H_2SO_4. Also other results will be discussed.

P6.2

ATMOSPHERIC CONDITIONS, REFLECTED IN CHEMICAL COMPONENTS IN SNOW OVER EAST QUEEN MAUD LAND, ANTARCTICA

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The chemical components in snow reflect the atmospheric environments. At several locations in the East Queen Maud Land, Antarctica, we obtained the detailed vertical distributions of hydrogen peroxide in surface snow layers. Taking the regional characteristics of chemical components in the snow into consideration, we can conclude that the snow in the inland high plateau reflects the atmospheric conditions in high altitude.

The concentrations of hydrogen peroxide were obtained at three different locations. It was clearly observed that the concentrations of hydrogen peroxide show periodic change and that the higher concentration occurs in the inland high plateau. The high concentration of chloride and nitrate, low pH level and high artificial radionulcides are also observed in the snow in the area. All the information suggests that the snow in the inland high plateau reflects the atmospheric conditions in high latitude. To trace the atmospheric environments into the snow deposits, we need more information on the chemical and physical metamorphic processes of the snow. The profiles of the oxygen isotope ratios do not directly agree with those of hydrogen peroxide in the region.

6.2

CHEMISTRY OF AEROSOL AND SNOW IN MIZUHO PLATEAU, ANTARCTICA

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The contribution of atmospheric aerosols to chemical composition of Antarctic snow was studied. Aerosol samples were collected in December, 1988, in Mizuho Plateau for the first time by the use of a modified Andersen Air Sampler, which separates aerosol samples into 12 size fractions ranging from more than 12 to 0.06-μm diameter. Drifting snow and pit snow were collected along the route from near the coast to 700-km inland.

The results on drifting snow clearly indicate that marine aerosol components (Na, Cl, SO_4, etc.) decrease with increasing distance from the sea coast inland and reach the lowest level at about 200 km from the sea coast. And then, SO_4, MSA (methanesulfonic acid) and NO_3 increase inland and suggest the presence of another source.

The size distribution of SO_4 and MSA in the inland aerosols have predominant peaks at 0.2-0.3 μm diameter, and run parallel indicating they are closely related or of the same origin. NO_3 shows a quite different size distribution from those of SO_4 and MSA and the origin of NO_3 seems to differ from these.

Na has its dominant peak around 1 μm and shows no evidence of marine origin. The calculation of nss-SO_4 referring to gross Na does not make sense for Antarctic inland aerosols.

I.3

MODEL ESTIMATES OF ANTARCTIC TROPOSPHERIC CHEMISTRY PERTURBATIONS DUE TO THE "OZONE HOLE" PHENOMENON

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Tropospheric trace gas concentrations for Antarctic conditions are obtained by the use of a photochemical model that describes the nitrogen, oxygen, hydrogen and carbon cycles of ozone destruction. The influences of total ozone depletion on tropospheric ozone, carbon monoxide, hydroxyl and some other constituents, including their diurnal variations, are investigated. Model results are compared with experimental data obtained at Mirny Station and aboard the research ship *Polarstern* in the Weddell Sea during 1989-1990.

PI.8

SUDDEN PHASE CHANGE IN OZONE QBO

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Quasi-biennial oscillation of total ozone has been studied over different latitudes using Nimbus 7 SBUV data. The variation of ozone over 10°N shows a missing QBO peak around 1983-1984. For the equatorial belt the peak during this period is definitely less pronounced than the ones preceding and following it. At 20° the phase of QBO is shifted so that it inherently attains a minimum in this period.

A detail heightwise study of ozone QBO using satellite measurements of O_3 mixing ratio is done along 75°E longitude. This shows some sudden phase changes in QBO in the altitude range of 30-40 km. Corresponding to the missing QBO peak over 10°N the layers above 34 km show positive peak whereas those below attain a negative value. For 0° latitude below 34 km the QBO oscillation is negative and below 31 km it shows a negative peak. For 20°N there is a minimum at that period which becomes more pronounced below 34 km. All this is indicative of some disturbance during 1983-84 which affected ozone distribution up to 35 km. The effect of El Chichon eruption in 1982 cannot be ruled out (Angell, 1990, private communication). The volcanic dust thrown up is supposed to have penetrated up to 35-km altitude. This being a preliminary study will lead to elucidate the propagation and effect of volcanic dust in the ozone layer around the globe.

P2.10

VOLCANIC EMISSIONS FROM MOUNT EREBUS, ROSS ISLAND, ANTARCTICA

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Mt. Erebus on Ross Island in the southern Ross Sea is an active volcano which contains a persistent convective lava lake of anorthoclase phonolite magma. Degassing of the magma causes a major volcanic gas plume which contributes significant quantities of volcanic aerosols to the troposphere in the Ross Sea area.

SO_2 emission rates have been measured annually since 1983 by correlation spectrometer (COSPEC V). In 1983 SO_2 emissions were 230 Mg/day but declined dramatically to 20 Mg/day in 1984 during a three-month period of sustained strombolian eruptive activity, ejecta from which buried the lava lake. Since 1985 emission rates have shown a general increase reaching 70±20 Mg/day in January 1991. The SO_2 emission is a function of the surface area of exposed magma in the summit crater.

Filter packs consisting of a 1-2 micron PTFE particulate filter followed by 2 to 4 filters treated with $^{7}LiOH$ or TBAH have been used to examine the nature and composition of particulate and acid gas aerosols. Using element/sulfur ratios measured on the filters and the COSPEC SO_2 emission rates, estimates have been made of emission rates for over 25 elements. Filters collected in 1986 had Cl/S=10-20 and F/S = 5-10 which implies emission rates for Cl and F of 200-1200 and 100-500 Mg/day, respectively.

Mt. Erebus is a major and significant source of aerosols to the troposphere in the southern Ross Sea area. The residence time and dispersal of these aerosols are poorly known at this time, but it is possible that Erebus may be a source of inorganic HCl found in snow and ice cores from East Antarctica.

P6.7

ESTIMATES OF ICE/SNOW COMPOSITION IN RELATION TO TROPOSPHERIC CHEMISTRY IN ANTARCTIC ATMOSPHERE

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During the sixth Indian Scientific Expedition to Antarctica (26 November 1986 - 22 March 1987), a few ice/snow samples at locations near Maitri station (Indian station in Schirmacher Oasis) in east Antarctica were extruded from the surface and at various depths below the ice/snow cap during the period from 19 January to 4 February, 1987. These samples were analyzed for radicals Cl^-, SO_4^{2-}, NO_3^- and NH_4^+ with Spectronic 2000 calorimeter and for Na^+, K^+ Ca^{2+} and Mg^{2+} with double beam Atomic Absorption Spectrometer. Before the chemical analysis, each sample was filtered through Whatman 41 cellulose filters. Maximum variation (76.1 times) was noticed for NH_4 followed by K^+ (18.1 times) and minimum for Ca^{2+} (2.3 times). It is difficult to identify how the snow/ice samples acquired various ions in such a range of concentrations for ion concentrations depend not only on the location and strength of pollutants but also on the meteorological conditions. The ratio of total anions to total cations can be used as an index of acidity or alkalinity. If the ratio is more than unity, it is considered alkaline in nature. These ratio values for our snow/ice samples varied from 0.499 to 3.064 with an average ratio value of 1.176 (acidic). This analysis revealed that old snow samples (collected from various depths) are dominantly alkaline while fresh snow samples (top of polar cap) are acidic in nature indicating thereby that anthropogenic air pollution, possibly due to long-range transport, has an increasing impact in recent years as compared to the past in Antarctica.

Clean air facility, first season.

4.3

NEW CHEMICAL STRATIGRAPHY FOR BYRD STATION, ANTARCTICA

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A 164-m deep, 10-cm diameter ice core, was obtained at Byrd Station Surface Camp (NBY-89), West Antarctica in November 1989. In addition, two 25-m deep ice cores were recovered at 5-km and 9-km distances upstream from the main core; 2-m deep pits were dug at each drilling location.

This study grew out of the perceived need to establish a new and updated chemical stratigraphy record from the present snow surface which would tie into and overlap the longer climate and environmental records available from the analyses made on the deep ice core obtained at this location 21 years earlier. (All studies on the deep ice core began with the index horizon 90 m below the 1968 surface.) The full chemistry study program includes oxygen and deuterium isotopes; acidity (ECM); H_2O_2, CO_2, $^{13}C/^{12}C$, CH_4, N_2O, O_2, Ar and N_2; INAA; volcanic dust; morphology and chemistry of particles which will be reported elsewhere. This paper is limited to the results obtained by ion chromatograph measurements (Dionex 4040) for major anions and cations, as well as $HCOO^-$ and $CH_3SO_3^-$ made in a clean room laboratory.

Seasonal variations of $CH_3SO_3^-$ concentrations in the pit samples show summer to fall maxima similar to summer maxima of NO_3^- and SO_4^{2-}. Below the pore close-off depth $HCOO^-$ concentration levels are 1 to 2 ng/g and nearly constant. Long-term multi-year average $CH_3SO_3^-$ concentration levels range from 2 to 10 ng/g and show a clear average depression of about 2 ng/g during the 18th and 19th centuries associated with a slight increase in the $\delta^{18}O$ values. The nearly 1000-year records contained in the main core (based on $\delta^{18}O$ and ECM) showed no significant long-term trends in concentration levels of Cl^-, NO_3^-, SO_4^{2-}, Na^+ and Mg^{2+}. Most measurements for NH_4^+, K^+ and Ca^{2+} were below the 1 to 4 ng/g detection limits of our instrument. Several excess SO_4^{2-} peaks in the main core are identified as being of volcanic origin and all are correlated with deep ice core layers from Byrd Station and Camp Century, Dye 3, Milcent and Crete, Greenland.

P6.6

PHYSICAL PROCESSES AFFECTING POLAR SNOW CHEMISTRY

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The Antarctic ice sheet preserves information on past changes in environmental conditions in the form of variations in both the physical and chemical character of stratigraphic features. A 164-m deep ice core was recovered at Byrd Station Surface Camp (NBY-89), West Antarctica in November 1989. The core was obtained to perform a detailed and integrated multi-parameter environmental analysis on the approximately ten-century chronology represented by the core, and to overlap and extend to the surface the numerous paleorecords acquired from the analyses of the 1968 deep ice core. Physical property measurements were made to investigate conditions of surface snow deposition, summer surface melting, snow/firn densification and air bubble enclosure processes. Diurnal, seasonal and annual variability in the physical environment at the surface affects snow chemistry through changes in the snow accumulation rate, and the near-surface water content, the permeable air-channel structure and the band width of the age distribution for formation of air bubbles. Continuous bulk density and visual stratigraphy measurements were made in a field trench. Detailed laboratory experiments were performed at eleven depth levels for crystal size, c-axis orientation, total air content and ultrasonic wave velocity. Pore close-off data was obtained over the permeable layers by continuous air-flow measurements with a resolution of 1 cm. Alternate layering of permeable and impermeable bands were measured at the 49.5- and 56.5-m depths. This layering structure is the result of seasonal variations in the original snow density which formed near the surface during short-term alterations in climate. Depth hoar layers observed suggest the existence of water vapor transport from spring/summer layer to fall/winter layer by molecular diffusion. This net flow of water molecules and associated dilution effect appears to cause irregularities or aberrations in the oxygen isotope record. The same effect is the probable cause of the enhancement or changeable sequences in the impurity concentration distributions, especially observed in firn stratigraphy older than 4 or 5 years.

P2.14

THE MEASUREMENT OF RADIOACTIVITY IN THE ANTARCTIC ATMOSPHERE

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For the past 28 years, the Environmental Measurements Laboratory has measured radioactivity on air filter samples collected at the global network of sites which constitute the Surface Air Sampling Program (SASP). Logistical problems with some remote sites, plus the relatively short half-life of ^7Be, the most prominent radioisotope that we measure, were two important factors that led to the development of the Remote Atmospheric Measurements System (RAMS). During 1990, RAMS were installed at the Mawson, Palmer and Marsh Stations in Antarctica.

RAMS are used to measure the gamma-ray activity on air filter samples at the field locations using a NaI(Tl) detector and to transmit the resulting spectra to EML using the Argos communications system flow aboard NOAA satellites. The resulting spectra are unfolded using linear least-square analysis software on our central laboratory computer. This provides almost real-time communication and analysis of data from remote areas. The current RAMS consists of a shielded 5" x 4" diameter integral line NaI(Tl) detector coupled to a multi-channel analyzer. The shield and detector are set up in a thermally controlled environmental box. The multi-channel analyzer is controlled by a lap-top computer which formats the data, sends the data to a transmitter and records it on a floppy disk. A power supply and transformer are also required for continual DC operation. Preliminary data from May 1990 through January 1991 indicate that the ^7Be concentrations at Palmer and Marsh may range from about 5 to 50 fCi/m^3 while the concentrations at Mawson are considerably higher and may range from about 50 to 150 fCi/m^3.

Building interim clean air facility, January 1975, New Pole Station. (Komhyr, Oltmanns and Zoller).

5A.3

APPLICATION OF AN ATMOSPHERIC TRACER MODEL TO THE HIGH SOUTHERN LATITUDES

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A diagnostic tracer model has been developed based on the University of Melbourne General Circulation Model (GCM). This model is a spectral rhomboidal wavenumber 21 with nine levels in the vertical. The travel model uses as input the three-dimensional distribution of winds simulated in a complete annual cycle integration of the GCM. The GCM produces a realistic climatology of the high southern latitudes (including baroclinicity and variability) which is important for its application to the present problem of realistic modelling of atmospheric dispersal in the Antarctic region. The tracer model includes, among other features, the effects of vertical and horizontal diffusion and moist and dry convection.

The model has been applied mainly to testing the sensitivity of the carbon cycle to various changes, with particular reference to the high southern latitudes and the Antarctic region. Some of the results presented focus on the interpretation of idealized experiments, in which sources and sinks of CO_2 are prescribed in simple ways. We examine the time scale of the transmission of the tracer from the Northern Hemisphere to the Antarctic region and compare this with that implied from measured data. We also determine these times in the case of sources in the midlatitudes of continents of the Southern Hemisphere. A further experiment was conducted in which a cloud of material was placed over the Antarctic continent and we examine the nature of its dispersal to the north, across the sub-Antarctic trough and into the westerlies.

Preliminary results will be presented of computer runs designed to simulate the atmospheric carbon cycle with realistic distributions of sources and sinks. The sources are taken from published work and the formulation of the source and sink terms over oceanic areas will be discussed. Results to be presented will indicate the sensitivity of the annual cycle of CO_2 at Antarctic stations to the precise formulation of these sources and sinks over the expansive southern oceans.

4.1

INSIGHT INTO THE SULFUR AND NITROGEN BIOGEOCHEMICAL CYCLES GAINED FROM GLACIOCHEMICAL STUDIES IN ANTARCTICA

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Comprehensive glaciochemical studies have been conducted at several Antarctic locations on the Antarctic Plateau (South Pole, Vostok) and in more coastal regions. Determination of all major ions (Na, NH_4, K, Mg, Ca, H, F, CH_3COO, HCOO, CH_3SO_3, Cl, NO_3 and SO_4) has provided an in-depth understanding of the chemical composition of present and past Antarctic precipitation. After a brief overview of spatio-temporal variations of major species we will focus on what we have learned from such studies on the sulfur and nitrogen cycles in the high southern latitudes and their response to natural phenomena (volcanic eruption, ENSO event, great climatic change) or to anthropogenic emissions.

Glaciochemical study indicates that except after volcanic eruption of global concern (which enhances the sulfuric acid background level of Antarctic ice), the sulfate present in the Antarctic atmosphere is mainly marine (oxidation of dimethylsulfide, DMS, provided by the marine biota) in origin. Our data suggest that high latitude marine DMS emissions are strongly enhanced during years with strong ENSO events (1982, for instance). Our study of the Vostok ice core which covers the last climatic cycle reveals that the oceanic atmospheric sulfur cycle was also strongly modulated by the climatic conditions in the past.

NO_3 content of high latitude precipitation reveals no evidence (as sometimes argued in earlier studies) of a positive correlation with solar activity. That, therefore, suggests that NO_x production in the high atmosphere (upper stratosphere, mesosphere) does not contribute significantly to the Antarctic NO_3 budget. Long range transport in the upper troposphere as well as input from the lower stratosphere represents dominant NO_x sources supplying the Antarctic atmosphere. Our data suggest that the NO_3 budget of Antarctica is disturbed by volcanic eruption (S-N cycle interaction) and more recently by the ozone hole.

P1.12

IMPLICATIONS OF THE ISOTOPIC PATTERN OF ANTARCTIC CARBON DIOXIDE AND METHANE ON THE SOUTHERN HEMISPHERIC CARBON CYCLE

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Our recent understanding of the global atmospheric carbon budget is mainly based on the temporal and spatial distribution of the CO_2 and CH_4 concentrations in the atmosphere. Completely independent information, leading to important constraints of the geophysical cycles of these trace gases, can, however, be drawn from isotope observations. In this context, remote southern hemispheric sites play a particularly important role due to the relatively well defined and homogeneous source and sink distributions in this part of the globe.

A continuous record of high precision $^{14}CO_2$ observations from 1983 to 1989 at the German Antarctic Georg von Neumayer station (GVN, 70°S, 8°W) are presented. Including parallel atmospheric observations from the Cape Grim Baseline Station (41°S, 145°E) as well as radiocarbon measurements of the South Atlantic surface water these data can be used to parameterize the atmosphere/ocean CO_2 gas exchange rate with the southern oceans.

Weekly high-volume samples for atmospheric methane concentration and isotope (^{13}C, ^{14}C, ^{2}H) analyses have been collected at GVN from 1988 onwards. The concentration record shows a very regular seasonal cycle similar to what has been observed at other remote sites in mid and high latitudes of the southern hemisphere. This seasonality is discussed considering first measurements of methane isotopes as well as results from transects taken over the Atlantic ocean from 55°N to 70°S.

P1.18

OZONE MEASUREMENTS AT THE BRAZILIAN ANTARCTIC STATION

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Surface ozone measurements were made at the Ferraz Brazilian Station (62°05'S, 58°23'W). A complete seasonal cycle was obtained in 1989, and repeated in 1990. Minimum concentrations are observed in January, less than 10 ppbv (parts per billion by volume), and maximum concentration in July–August, when around 30 ppbv are observed. Ozone concentrations at Ferraz show generally slow variations with time. An exception to this is an abrupt decrease, on certain occasions, to values some 20 ppbv less during the winter period. These episodes seem to be related to abrupt changes of airmasses that are sampled at our site. The recovery to "normal" values may take between a few hours to several days. Ozone measurements in the whole troposphere have been started recently using ozonesondes launched on balloons. Measurements made in January–February 1991 show rather smooth ozone profiles compared to tropical ones. Typical partial pressures vary between 10–15 nanobar, nb, to about 20–25 nb in the higher troposphere. There is a sharp transition between ozone in the troposphere and stratosphere, at a pressure of between 400 and 300 millibar, mb, at which level the partial pressure of ozone increases stepwise by about 20–40 nb.

P5A.3

THE ROLE OF THE ACOUSTIC SOUNDER IN DEFINING THE STRUCTURE AND THE DYNAMICS OF THE ANTARCTIC BOUNDARY LAYER

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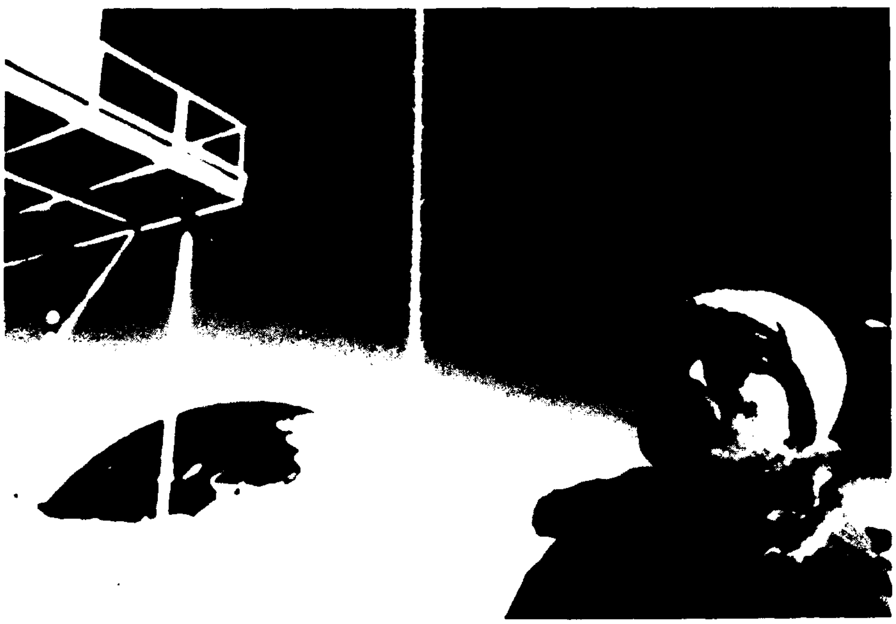
It is well known that the structure and the dynamics of the boundary layer play an important role in controlling tropospheric chemistry. This is particularly true for the Antarctic tropospheric regions where the vertical mixing is extremely damped because of the almost continuous presence of strong ground-based inversions. Beside this, the dynamics in the lower layer of atmosphere (50 - 1000 m), and the surface energy budget are other important phenomena to investigate because of their fundamental role in influencing the global climate. A sodar system seems to be an appropriate tool to give information on wind and turbulence fields with the level of accuracy we need in such studies. The facsimile representation of echo will also give an indication on the thermal structure of the PBL.

In three successive summer campaigns a sodar system was deployed in Antarctica by a group supported by the Antarctic Research Program. During the first and the second campaign (1986-1987 and 1987-1988) the system was installed on the Nancen ice sheet at the confluence of the Reeves and Priestly glaciers.

In this paper we present some examples of the phenomena observed: strong convection, subsidence, horizontal and wave structures, katabatic winds. Furthermore will be shown the results of some computations in which the vertical heat flux, the dissipation rate, and the dissipation length have been evaluated.



Riming of tethersonde lead, K. Kikuchi, balloon inflation shelter, New Pole Station, 1975.

P5B.2

AIR MASS EXCHANGE BETWEEN TROPOSPHERE AND STRATOSPHERE IN THE SOUTHERN HIGH LATITUDE BASED ON OBSERVATIONS OF LATITUDINAL-VERTICAL DISTRIBUTIONS OF OZONE IN 1987-1990

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The observations in the Southern Hemisphere indicate constant or slightly decreasing ozone levels. This fact suggests that most of tropospheric ozone in this hemisphere is coming from the stratosphere. Tropospheric ozone is an important species controlling the main process of atmospheric chemistry in this hemispheric troposphere.

In order to know the detail process of ozone transportation from stratosphere to Antarctic troposphere, the Japanese Antarctic Research Expedition has carried out observations of total ozone and vertical ozone profile on board the observation vessel *SHIRASE* every 5-degrees of latitude from the equatorial region to Antarctica in the period from mid-November through mid-December for four years.

Latitudinal-vertical cross sections of ozone clearly indicate the high ozone tongue from stratosphere to troposphere at around 30°S and the low ozone bank at around 60°S, strongly suggesting that ozone in the Antarctic troposphere is injected from stratosphere to troposphere in mid-latitude, and then transported into Antarctica through the upper troposphere.

P2.3

CARBONACEOUS AEROSOLS: MEASUREMENT AND SOURCE ATTRIBUTION

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Recent measurements of atmospheric particles at the South Pole show the presence of elemental carbon. This component of the total carbonaceous aerosol fraction is linked to combustion processes, and is considered to be an indicator of allochthonous (transported) carbon to the Antarctic environment. Unlike elemental carbon, organic particulate carbon has not been studied at the South Pole, and yet has been shown to be an important contributor to the total carbonaceous aerosol mass at other remote sites. Organic carbon particles scatter solar radiation (depending on size and composition); elemental carbon particles can scatter *and* absorb solar radiation. In addition, molecular species present as organic carbon aerosol are photochemically active and are involved in the production of atmospheric oxidants. So, in addition to affecting atmospheric optical properties, the presence of reactive organic aerosols may also influence photochemical processed occurring within the Antarctic atmosphere.

One approach used to study the atmospheric concentrations of particulate carbon involves development of a chemical species mass balance. Here, the relative proportions of elemental carbon to organic carbon aerosol particles are measured. This accounting provides useful information needed to describe the ambient levels of optically active carbonaceous aerosols (i.e., light scattering versus light absorbing species); these concentration data may be used further in predictive models of light extinction and radiative transfer in the Antarctic atmosphere. A further advantage to this carbonaceous aerosol mass balance is the ability to relate emission source type to concentrations of molecular tracers present as organic aerosol. Given this quantitative framework, it is possible to describe discrete sources of particulate carbon to the Antarctic region.

P4.4

GLACIO-CHEMICAL STUDIES IN THE RONNE AND EKSTROM ICE SHELF REGIONS

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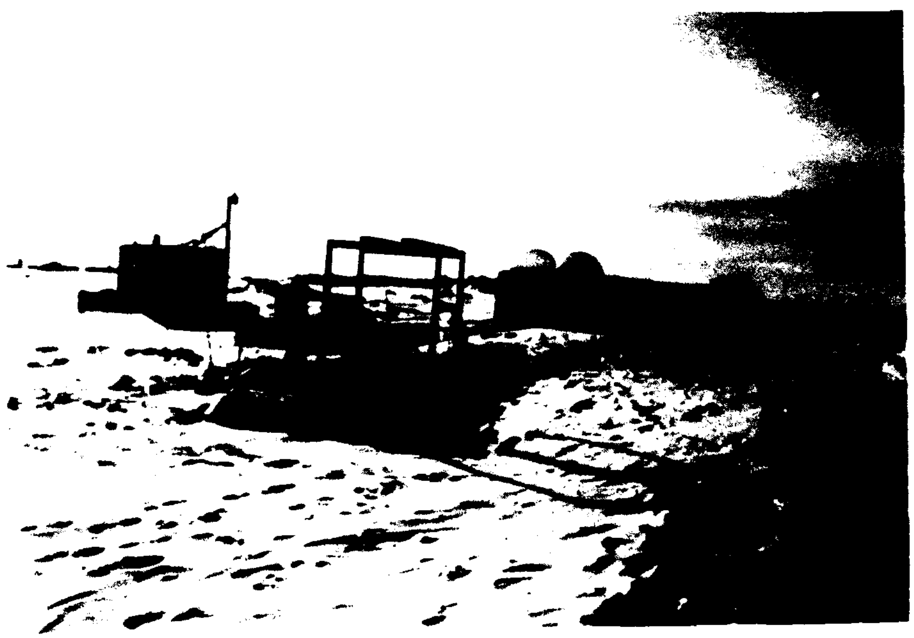
In contrast to central Antarctica, there are only a few glacio-chemical records available from the Antarctic ice shelves. Due to the simple topography, a relatively high accumulation rate, and the immediate access to the ocean, these snow fields are expected, however, to be particularly useful in deriving high resolution records of the coastal Antarctic aerosol body. To assess the potential of this archive properly for deep drilling efforts, basic information on the spatial and temporal pattern of snow impurities are gathered in the course of several glaciological surveys on the central Ronne Ice Shelf (55°W) as well as on the Ekström Ice Shelf and its catchment area (8°W). A series of snow pits and shallow ice cores are sampled for stable isotopes, major ions, MSA, and natural radio-isotopes on a 500-km scale, approximately. In addition, "mineral-acid proxy stratigraphies" are established by electrical conductivity profiling of firn cores, and a 200-m deep ice core from the Ronne Ice Shelf.

Opposite to the small Ekström region, the Ronne-Filchner area (including the Berkner Island summits) shows a very regular snow deposition in the range of 15-25 cm w.e. yr^{-1}. This easily allows for precise annual layer counting from the stable isotope, nss-sulfate, or MSA stratigraphy. In addition to the obvious continental effect in the spatial distribution of stable isotopes, snow accumulation rate and sea-salt, the mean snow concentrations of nss-sulfate and MSA exhibit a significant decrease with distance from the coast as well. No trend is observed, however, for nitrate concentrations.

Comparisons of firn profiles with long-term aerosol records at the coastal G. v. Neumayer station gives evidence for a higher scavenging ratio of total nitrate if compared to the (sub-micron) nss-sulfate, MSA, and radioactive aerosol components.



Solarimeter stand, inflation shelter, auroral observatory; New Pole Station in background, 1974.

4.2

THE RATIO OF MSA TO NON-SEA SALT SULFATE IN ANTARCTIC PENINSULA ICE CORES

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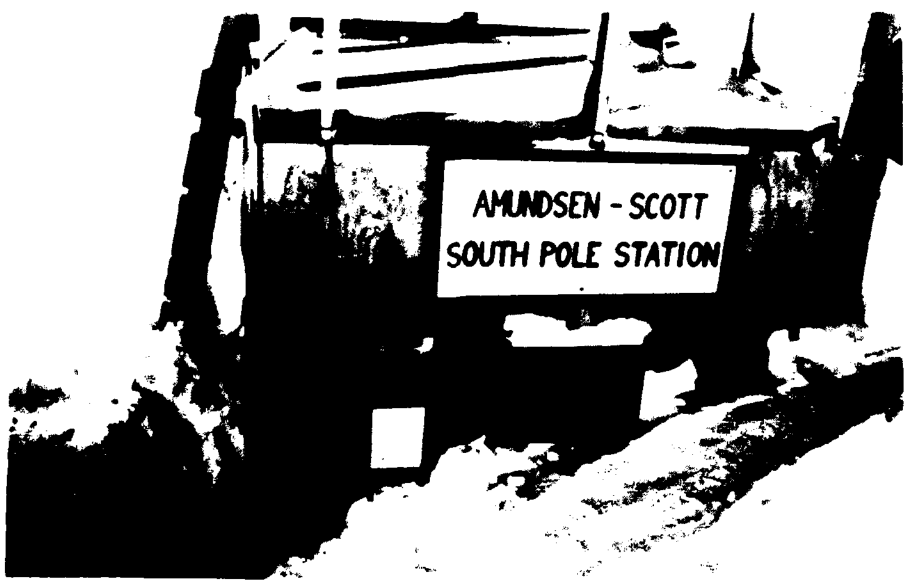
Methane sulfonic acid (MSA) measurements made on the Dolleman Island ice core (collected from a site on the west of the Peninsula at 70°35'S, 60°55'W, altitude 398 m) show significantly high concentrations (typically 100-200 ng/g) compared to values recorded in ice cores and snowfall from elsewhere in Antarctica. The MSA data from Dolleman Island are further unique in that they demonstrate a seasonal maximum in winter rather than summer when photochemical activity is greatest.

MSA is a breakdown product of the gaseous dimethyl sulfide (DMS) emitted by marine phytoplankton. The unusually high values from Dolleman Island could be explained by its close proximity to the biologically productive Weddell Sea; relatively high concentrations of other marine ions may support this. However, the consistent observation of well defined peaks in the winter is more difficult to interpret. The decomposition of DMS can proceed by two reaction pathways which produce either sulfuric acid or MSA as end products. In contrast to the behavior of MSA, it is a characteristic of Dolleman Island that the concentration of sulfate not attributable to a marine salt source (i.e., non-sea salt sulfate) is both high, and peaks in the summer.

This paper demonstrates the seasonal pattern of deposition of MSA and of non-sea salt sulfate from Dolleman Island. We consider whether the Dolleman results are representative of the wider region by comparison with similar data from two other Antarctic Peninsula ice cores: one collected from a site on the spine of the Peninsula at a similar latitude but 150-km west (70°31'S, 65°01'W, altitude 1900 m) and another from Gomez Nunatak (74°01'S, 70°38'W, altitude 1130 m), also on the spine but 500 km further south.



Head of Holy Stairs; Old Pole Station about 40 feet below.

P5A.4

THE FRACTIONATION OF SEA SALT DURING TRANSPORT ACROSS AN ANTARCTIC ICE SHELF

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Recent analyses of ice cores have shown that ionic species of marine sea salt origin often differ from bulk sea water ratios. This paper examines whether this phenomenon can be explained by progressive fractionation during transport from open water to the deposition site.

In January 1990 a series of surface snow samples was collected along a 120-km traverse of the Fimbul Ice Shelf in Dronning Maud Land. At the first site, 7 km from the ice shelf front (69°55.6'S, 0°06.8'W), and at the last site (70°58.7'S, 0°12.1'E), shallow pits were dug to allow sampling through about one meter of snowfall at a resolution of about 30 mm. Between these two sites, four replicate samples of the surface layer (to a depth of about 20 mm) were collected every 4 km. At both pits and surface sites, samples were scraped directly into pre-cleaned plastic accuvettes, and shipped in cold storage back to the UK for analysis. Precautions were taken to avoid contamination of the samples in the field, including wearing disposable polythene gloves and ensuring the sample area was upwind of the party.

Each sample has been analyzed by ion chromatography for chloride, nitrate, sulfate, sodium, potassium, magnesium and calcium. Results from the replicate surface samples show very little local variability, whereas the trends from the ice shelf front are very clear. For example, chloride increased from the ice shelf front to a maximum value approximately 40-km inland, before decreasing rapidly to a value of 10% of the maximum by 120 km inland. Sulfate followed a similar trend to 40 km, but did not subsequently decrease by such a large factor. This result may be due to the presence of a large amount of sulfate derived from marine biogenic activity rather than from a seal salt source. It does show that sea salt may be washed out of the atmosphere earlier than biogenic sulfate. Results from the analysis of the cations will also be presented, and the effects of aerosol fractionation discussed.

A further intriguing result was the relatively high concentrations of nitrate in all of the surface snow samples in comparison to the pit samples. This may be an indication of a post-depositional loss of nitrate from the snow surface.

1.2

AIRCRAFT MEASUREMENTS OF THE TROPOSPHERIC OZONE CONCENTRATION OVER THE ANTARCTIC REGION

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Aircraft measurements of the tropospheric O_3 concentration were made over Syowa Station (69°00'S, 39°35'E), Antarctica from May 1989 to January 1990, except during the polar night months of June and July. The O_3 concentration increased with height for the whole period of the measurements. The lower tropospheric O_3 showed a prominent seasonal variation with maximum concentration in winter and minimum concentration in summer, which is very close to the result of continuous O_3 measurements initiated at Syowa Station in February 1988. In the upper troposphere, the concentration reached high values in winter, decreased gradually until early in November, and then increased again. As a result, the height-dependent difference of the concentration was reduced from late in autumn to early in spring and enhanced in remaining seasons of the year. The seasonal variation of the upper tropospheric O_3 was similar to that of the lower stratospheric O_3 concentration measured by ozonesondes and that of the total amount of O_3 by a Dobson spectrophotometer over Syowa Station. It was also shown, from the results of ground-based O_3 measurements at the South Pole, Syowa Station and Cape Grim, Tasmania, that the concentration is always higher in high latitudes than in middle latitudes of the southern hemisphere, and appearance of the minimum concentration of the seasonal variation is delayed by almost one month at the South Pole more than at Syowa Station and Cape Grim.

These results may suggest that slow downward transport of O_3 from the stratosphere through the troposphere occurs in the Antarctic region throughout the year, and the air with low O_3 concentration is transported from southern middle latitudes into the Antarctic region through the lower troposphere from spring to early in autumn.

P5A.2

VERNAL ATMOSPHERIC MIXING IN THE ANTARCTIC

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Aerosol concentration, ozone concentration and meteorological parameters were measured at McMurdo and South Pole Stations during a spring storm, which reached the Antarctic interior. Nacreous clouds were sighted preceding the storm indicative of stratosphere flow from lower latitudes. These measurements and observations, along with upper air and surface analysis indicate that vigorous tropospheric/stratospheric exchange of air occurs near 75°S during the spring.

The elemental composition of collected aerosol changed coincidently with different stages of the storm. During the storm event in September 1983, surface ozone concentration varied from 20 to more than 100 ppbv at McMurdo, but remained less than 20 ppbv at the South Pole indicating that deep mixing which occurred at the periphery of Antarctica during the spring storm did not continue over the interior of the continent. The warm marine air associated with the spring coastal storm infiltrated the interior of Antarctica including the Polar Plateau, producing a record surface temperature and an aerosol concentration twice the September mean. This system was unusual as the warm front apparently reached the surface of the South Pole.

Crustal material was transported to the periphery of Antarctica through the upper troposphere or lower stratosphere. Enhanced aerosol concentration was transported to the South Pole through the lower troposphere. Vigorous exchange occurred at latitudes of greater than 78°S, which probably exchanged both marine aerosol and water vapor into the lower stratosphere.

Zoller and Rasmussen on way to Three Mile Station, 1975.

P5A.5

RELATIONSHIP BETWEEN SOUTHERN HEMISPHERE PLANETARY WAVE ACTIVITY AND OZONE LEVELS

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Gridded data from the European Center for Medium Range Weather Forecasts and NIMBUS 7 TOMS data are used to isolate large scale atmospheric wave activity in the Southern Hemisphere and relate these to observed ozone levels. The last decade has witnessed an alarming decreasing trend of total ozone over Antarctica with pronounced year-to-year variations. Low values during October have been related to low 100-mb temperatures over Antarctica, a strengthened polar vortex, and the westerly phase of the quasi-biennial circulation[1,2]. These relationships not only correctly explained the 1987 ozone minimum that occurred in conjunction with the westerly maximum of the QBO but appear also to agree with recovered levels of ozone during 1988. Our presentation explores these relationships for the period 1986-1989, quantifying interannual and intraseasonal variability of ozone and relating these to the changes of selected fields in isentropic coordinates. The horizontal and vertical distribution of Isentropic Potential Vorticity (IPV) as a function of season is presented, and salient features of upper-level IPV are linked with observed changes in total ozone. Though the gridded data is only available up to 100 mb, the analyses permit identification of cases with a dominant role of lateral mixing in total ozone content which is mostly due to changes in the 12-20 km layer[3].

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P2.6

HYGROSCOPIC AEROSOLS AT THE SOUTH POLE

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Aerosols were sampled at the South Pole during the austral summer of 1989-1990, using a low-pressure impactor. The low pressure impactor was able to collect all aerosols larger than 0.01 μm in diameter in minutes. Sampling substrates used were electron microscope grids overcoated with plain collodion, with $BaCl_2$, Ca, or Nitron to detect chemical composition of the aerosols. We used a scanning transmission electron microscope (STEM) equipped with an energy-dispersive X-ray analyzer, which can detect elements with atomic numbers greater than 6 (Carbon) for individual particles.

Almost all particles were hygroscopic and were probably droplets of H_2SO_4. These results confirmed Ohtake's previous observations. Ninety-nine percent of the aerosols, regardless of their size, were pure sulfuric acid. Concentration of Aitken condensation nuclei were approximately 150/cc according to GMCC data at the South Pole. The samplings were made at the Clean Air Facility, which is located upwind and free from any possible local contamination at the South Pole station.

Since stratospheric aerosols are probably dominated by H_2SO_4 all over the planet earth (6), it is not surprising that we are finding the same at the South Pole.

We surmise that the mechanisms of H_2SO_4 aerosol formation may be as follows:

1. Oxidation of DMS (CH_3–S–CH_3),
2. Volcanic activity, and
3. Air pollution, probably through SO_2 by combustion of coal and oil.

The Antarctic and stratospheric aerosols basically do not contain a very large fraction of man-made pollution particles or volcanic dusts as Aitken condensation nuclei: they are essentially all H_2SO_4. We tentatively conclude that the H_2SO_4 aerosols found in the Antarctic are formed by oxidation of DMS, while stratospheric H_2SO_4 are more likely caused by volcanic activities.

Since H_2SO_4 aqueous solution could act as freezing nuclei for clear sky precipitation in the polar zones, cirrus clouds, and polar stratospheric clouds, laboratory experiments determining its freezing points as a function of dilution rates and size of solution are being conducted.

P1.3

TROPOSPHERIC OZONE OVER ANTARCTICA

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The dramatic depletion of stratospheric ozone in the spring over Antarctica that has taken place over the past decade has focused attention on the ozone layer in the Antarctic. Significant changes have evidently taken place in the troposphere as well. At South Pole the near-surface ozone concentration during the summer (D-J-F) has declined by nearly 20%. In recent years as part of intensive studies of the Antarctic stratosphere, vertical profiles of ozone have been obtained at several locations in Antarctica. Although these ozonesonde records are not of sufficient length to assess long-term changes, they do provide an opportunity for determining the representativeness of the South Pole record.

At locations throughout the Antarctic, the seasonal pattern in the troposphere appears to be remarkably similar to that at South Pole. The ozone concentration builds up during the austral autumn and remains near the maximum values through the winter. Day-to-day changes tend to be relatively small during this period. In the spring, ozone amounts become more variable with episodes of depleted ozone. This leads to the summer seasonal minimum. The continent-wide nature of seasonal ozone changes suggests that the tropospheric ozone distribution is driven by the seasonal change in solar radiation and large-scale circulation patterns.

P6.1

SOURCES OF CONTAMINANTS IN ANTARCTIC SNOW

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Snow-pack samples were collected at eight sites in Antarctica during the summer of 1988-89 to evaluate effects of the Erebus volcano on deposition. Elemental concentrations were determined by Instrumental Neutron Activation Analysis (INAA). Sources of elements in snow samples were investigated during receptor modeling from chemical profiles for the Erebus plume and magma, seasalt, and crustal material. Source contributions were estimated from least-squares fits of elemental concentrations in ambient samples to those in source emissions. Of the elements measured, arsenic and gold were highly enriched in the Erebus plume. Most of the samples had major contributions from crustal sources. However, Erebus was the most important contributor at the Erebus site, and contributed significantly to samples from Bird Saddle, Cape Crozier, the Fang Glacier, and at two snow depths at Terra Nova Saddle. No Erebus contributions were estimated for samples from the South Pole or the Newall Glacier. These results suggest that receptor modeling may be useful in determining the relative roles of sources of aerosol and gaseous precursors in Antarctica. Future studies of this kind should focus on characterizing a more aged Erebus plume, an anthropogenic background, and more realistic emissions profiles for local marine and crustal sources. Secondary species such as nitrate, sulfate, and methane sulfonic acid would help resolve sources and chemical transformations affecting atmospheric deposition in Antarctica.

PI.16

GASEOUS NITRIC ACID AND AMMONIA AND PARTICULATE AMMONIUM NITRATE IN THE MARINE ATMOSPHERE OF THE WEDDELL SEA IN NOVEMBER/DECEMBER 1990

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During a recent expedition (November 18 to December 17, 1990) on board the German research vessel *Polarstern* from Punta Arenas (Chile) to Georg-von-Neumeyer station at Akta Bay we measured the concentrations of gaseous nitric acid and ammonia by a laser-photolysis fragment-fluorescence (LPFF) method. The measurements were performed continuously with a detection limit of 20 pptv for HNO_3 and 150 pptv for NH_3 using a signal accumulation time of 1 h.

Furthermore, air samples were taken through cold and heated denuder tubes for quantitative analyses of gaseous nitric acid and particulate ammonium nitrate, respectively. These samples are being evaluated at present by ion chromatography at the home laboratory.

Sampling sites for both methods was the front of the ship's upper deck, 20 m above sea level. The denuder samples were taken at the radar tower of the ship, 28 m above the water surface. The HNO_3 sampling line was equipped with a salt separator to eliminate the influence of sea salt.

Because these measurements were performed quite recently, they are not yet analyzed at the time of the writing of this abstract. However, a first look at the results of the cruise from Cape Horn to Cape Norwegia (easterly direction) seems to indicate the decreasing influence of air masses from the South American continent on the HNO_3 and NH_3 concentrations. Close to settlements and scientific stations, power generation by fossil fuel clearly increased the atmospheric level of HNO_3.

Acknowledgement: This work was financially supported by BMFT, grant No. 0743131/2. We thank R. Fagentzer for the preparation and the chemical analysis of the denuder tubes.

P.13

^{222}Rn TO ^{220}Rn ISOTOPIC RATIOS IN THE ANTARCTIC TROPOSPHERE: PRELIMINARY RESULTS

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Atmospheric radon is one of the most reliable tracers to distinguish between continental and oceanic air masses in Antarctica due to its relatively simple atmospheric cycle. Nevertheless, in coastal stations the local production of radon by the exposed regolith and rocks cannot be neglected, mainly during summer when important ice melt occurs. A new detection method for radon is being employed routinely at the Brazilian Antarctic station "Ferraz" (62°W, 58°S) which is capable of discriminating local from imported radon based on the large difference between the half-lives of its isotopes. Results from previous whole-radon data collected since 1986 showed strong surges of radon in association with the passage of deep cyclonic circulation through the Drake passage. However, the use of this new technique during 1990 clearly demonstrated that not all of these surges originated from radon imported from the South American continent but were local contributions from emanations due to soil temperature increases. Subtraction of this local source revealed the true oceanic nature of these air masses. Matching radon data obtained from Ferraz with that obtained from a twin equipment installed in Punta Arenas (53°S, 71°W) is being made in order to determine transit times of the continental air masses.

Grant No. 9586, Brazilian Antarctic Program.

2.5

NITROGEN AND SULFUR SPECIES IN AEROSOLS AND THEIR RELATIONSHIP TO NATURAL RADIONUCLIDES AT MAWSON, PALMER AND KING GEORGE ISLAND

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A high volume aerosol sampling program was initiated at Mawson, Antarctica (67°36′S, 62°53′E) in February 1987. Samples are collected continuously for week-long periods and they are periodically returned to our laboratory for analysis. We extract a portion of the filters and measure the concentration of NO_3^-, $SO_4^=$, Cl^-, methanesulfonate (MSA), Na^+ and NH_4^+. A portion of each filter is measured for the natural radionuclides ^7Be and ^{210}Pb. The data for 1987 and 1988 (Savoie et al., 1991) show a very strong seasonal cycle in the aerosol concentration of nss–$SO_4^=$ and MSA with the maximum occurring in the Austral summer when oceanic primary productivity is at its peak. This cycle suggests that the oceanic source of MSA plays an extremely important role in the atmospheric sulfur budget in this region. The seasonal cycle for NH_4^+ is quite similar to that for MSA, perhaps indicating an oceanic source for this species as well. There is a pronounced seasonal cycle for aerosol NO_3^- but it is markedly different from that of the other species. The radionuclide data suggest that the NO_3^- (and 25% of the nss–$SO_4^=$) is most likely derived from continental sources as opposed to stratospheric or biogenic sources.

In early 1990, we established two additional aerosol sampling stations in the Antarctic: Palmer Station (64°46′S; 64°3′W); King George Island (62°11′S; 58°18′W). In this report we will present the results of the data obtained from the analysis of samples obtained concurrently at all three stations. The synoptic nature of the data set provides further information on the character and the distribution of the sources of these species and about the factors that affect the transport of these materials over the Antarctic continent.

P2.5

OPTICAL AND AEROSOL PARAMETERS OF THE ATMOSPHERE FOR ANTARCTIC COASTAL STATIONS

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Results of field measurements of integral transparency coefficient, spectral aerosol atmospheric optical depth (wavelength range from 0.4 to 1 μm) and the size distribution of aerosol particles larger than 0.4 μm in the surface layer of the atmosphere at Mirny station. The impact of volcano eruptions (e.g., Agung and El Chichon) on aerosol extinction of the solar radiation is estimated.

3.6

ANTARCTIC SULFATE: EREBUS CONSIDERED

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While a significant fraction of the Antarctic sulfate budget that is surface deposited is doubtless of marine origin, a review of airborne measurements of the flux of SO_2 from Mt. Erebus volcano continues to suggest an important volcanic contribution. Mt. Erebus is a 3794-m composite volcano on Ross Island, Antarctica (77.55°S, 167.17°E) and is the principal volcanic source in Antarctica. The crater of Mt. Erebus is persistently filled with magma and numerous active fumaroles and vents are found throughout the crater area. Noted for its lava lake and long periods of minor strombolian eruptive activity, Ross observed its extensive plume in 1841 as did Scott in 1903. Its lava lake has been present since at least 1972 and may well have been present for a far longer interval.

Airborne measurements of its SO_2 emission flux in November of 1980 ranged from 35-155 tonnes per day (76 ave)[1] and in December of 1983 from 110-440 tonnes per day (230 ave)[2]. Since activity at Erebus has increased since 1984 these emission values may be representative of longer-term emissions.

Regardless of which of the estimates in the literature are used for the sulfate flux to the Antarctic surface, this airborne work shows that the contribution of Erebus is potentially a very significant fraction of the total. In view of the geographical and meteorological setting of Mt. Erebus and the very slow conversion of SO_2 to $SO_4^=$ (expected at low light levels and cold temperatures) it is not unreasonable to expect Mt. Erebus to have a continental rather than merely local-scale impact.

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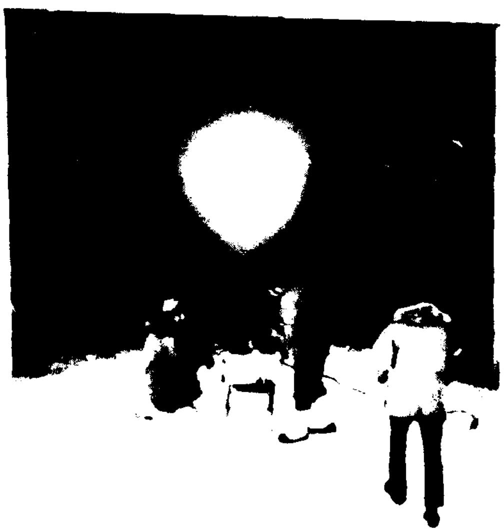

Hofmann and company preparing for a launch, 1975.

4.5

PARTICLES IN POLAR ICE AND THE INSOLUBLE BACKGROUND AEROSOL

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The insoluble background aerosol is an aerosol with a relatively long residence time and one that does not change over an extended region of the troposphere, possibly a whole hemisphere. The total aerosol consists of the background aerosol and a local aerosol which has a much shorter residence time and varies from place to place. In general, we expect that the local aerosol will tend to increase the relative population of the larger particles in the troposphere.

We have measured the size distribution (averaged over one or more years) of insoluble particles recovered from polar ice cores and related the results to the insoluble background aerosol[1,2]. Our measurements cover the optically interesting radius range 0.05-1.31 μm. To give a simple indication of size distribution, we divide the measured radius range into three subranges, "small" (0.05-0.13 μm), "medium" (0.13-0.38 μm), and "large" (0.38-1.31 μm) and specify a size distribution by the percentage of particles per gram of ice meltwater in each of the subranges. Analyzing these results and some new ones, we conclude that one can make a very good case for relating particles retrieved from Antarctic ice to the insoluble background aerosol, and that the size distribution of this aerosol has remained unchanged over the past 30,000 years. The invariance of the size distribution over such a long time tells us that we are, indeed, sizing an aged aerosol that has equilibrated in the atmosphere in such a way as to produce a unique size distribution which has lost any memory of its source and strictly reflects the aerodynamic equilibrium of the suspended particles. For Greenland ice, we have found some significant deviations in the size distributions which seem to reflect the influence of local aerosol sources. One of these deviations occurs in the holocene and can be attributed to a period of significant volcanism. The other deviations occur during the Wisconsinian glacial period and possibly reflect the influence of nearby continental sources under high wind conditions. Otherwise, all other holocene size distributions we have measured are consistent with an aged insoluble background aerosols similar to the one inferred from Antarctic ice.

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5B.2

TROPOSPHERE-STRATOSPHERE EXCHANGE OVER ANTARCTICA

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A number of Antarctic airborne air chemistry experiments were carried out in the 1976-1981 period using the NSF C-130 specially equipped for such research. The analysis of data from several of these programs provided some insights to mechanisms of exchange between the troposphere and the stratosphere. Exchange mechanisms which, apparently, can be examined with the available NSF Antarctic aircraft, include those associated with jet stream circulations and those resulting from terrain-induced unstable wave patterns. In these 1976-1981 studies, the Antarctic examples of stratospheric-tropospheric exchange were generally serendipitous results from more general programs rather than designed objectives, and many details of the exchange mechanisms cannot be described. This discussion presents a review of previous research studies, and how they might relate to current Antarctic problems in air chemistry. Possible future research programs for the specific examination of Antarctic exchange processes will also be discussed.

1.5

MEASUREMENTS OF LIGHT ORGANIC TRACE GASES IN ANTARCTICA

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Between 1982 and 1990 about 200 whole air samples were collected at the "Neumaier" Station in Antarctica (70°36'N and 8°22'W). These samples were analyzed by several gas-chromatographic techniques for a number of organic trace gases. In this presentation the results for light nonmethane hydrocarbons and methylchloride are presented. Yearly average mixing ratios were around 0.35-0.4 ppb for ethene and ethane, 0.01 ppb for acetylene, 0.2 ppb for propene, 0.07 ppb for propane, and 0.62 ppb for methylchloride. The mixing ratios of the heavier alkanes were in general below 0.01 ppb. The less-reactive trace gases, methylchloride, ethane, and acetylene, exhibit a systematic seasonal cycle with similar phases. The maxima are in late austral winter, minima in late summer. However, the relative seasonal changes differ, a few percent for methylchloride, about 50% for ethane and roughly a factor of 3 for acetylene. These cycles are in phase with the peak of biomass burning in the southern hemisphere, which is one of the major sources of these trace gases. But also the seasonal changes of their atmospheric photochemical removal rates can explain the observed seasonality.

1.4

SEASONAL AND SECULAR VARIATIONS OF TROPOSPHERIC OZONE IN ANTARCTICA

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Surface ozone at South Pole displays a distinct summer minimum with a significant downward trend over the period 1976 to 1990.

Since 1976, January-February ozone concentrations at South Pole have decreased by 17%. Over the same period, solar irradiance has decreased a similar amount concurrent with sky cover (cloudiness) which has increased about 20%. Data from coastal ozonesondes, air trajectory analyses, and model studies suggest that Antarctic tropospheric ozone is being destroyed in a classic (low NO_x) photolytic reaction due to increased UV penetration as a consequence of the stratospheric ozone hole. The observed tropospheric ozone decrease can be expected to result in an increase of about 4% in the biologically active UV radiation at the surface in coastal Antarctica.

If our results and hypotheses are supported by other researchers, we may have identified dramatic effects from the ozone hole on lower troposphere chemistry, and continent scale shifts in seasonal air flow patterns which began in the late 1970's.

2.3

THOUGHTS ON POSSIBLE CLIMATE MODULATION BY ARCTIC AND ANTARCTIC CLOUDS

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Clouds over the polar regions of the planet have unusual characteristics due to low temperature environment and perhaps in some cases to a lack of condensation or freezing nuclei. An additional interesting factor for climate change is that cloud cover occurs over a highly reflecting snow and ice surface. It seems probable that most nuclei that seed Antarctic clouds are natural, non-anthropogenic products, consisting either of sulfates or seasalts (in the case of cloud condensation nuclei) or clay (in the case of ice nuclei). We discuss how variations in these natural products might alter cloud microphysics, precipitation and climate and, in turn, to close the feedback, will ponder how these natural products might alter themselves, when or if climate changes.

P1.14

SURFACE OZONE MEASUREMENTS FROM DAKSHIN GANGOTRI, ANTARCTICA

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Surface ozone measurements for seven months were carried out in two spells from the Indian Antarctic station Dakshin Gangotri (70°05'S, 12°00'E) using the Indian electrochemical surface ozone recorder. Tropospheric ozone data from the Indian ozonesondes was also used in the analysis during the breaks in the continuous recording. The main objective of the measurements was to find out the nature of variation of surface ozone during the rapid depletion and fast recovery of total ozone in late winter and early spring.

The surface ozone values show a summer minima of the order of 20 nb and a winter maxima of the order of 42 nb. The transition from the low summer value to the high winter value occurs during June. From October to November there is a sharp fall in the surface ozone values. This sharp fall takes place at a time when the total ozone values are sharply rising from the lowest annual value.

Other notable features of the surface ozone at Dakshin Gangotri during 1989 are the near absence of diurnal variation even during the summer months and the absence of significant changes during blizzards. Results agree fairly well with the measurements reported from Syowa in Antarctica, a station located in nearly the same latitude. Comparison with a tropical station, Pune (18°32'N, 73°51'E), brings out the sharp differences in surface ozone distribution between the two stations. Pune has the highest values when Dakshin Gangotri has the lowest, and vice versa. The major difference, however, is that the systematic diurnal variation noticed at Pune almost round the year is not noticed at Dakshin Gangotri. The sharp changes noticed at Pune associated with different synoptic situations, break-up of inversion and wind changes are not noticed at Dakshin Gangotri.

P2.12

THE RADIATIVE EFFECTS OF CLOUDS IN ANTARCTICA

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An attempt has been made in this paper to discuss relevant inferences on radiation dispositional characteristics as modified by clouds through detailed examination of the observed data for two stations in the Antarctic region of which one is in the coastal area and the other is in the center of the elevated plateau region. Results obtained from sensitivity studies using a 2-D Zonally-Symmetric Climate Model with the objective of understanding the broad characteristics of cloud-radiative interaction in polar regions are also presented. Our findings indicate that the cloud-radiative interaction can lead to considerable changes in the thermal structure of the polar troposphere.

6.1

H_2O_2 AND HCHO CONCENTRATIONS IN POLAR SNOW AND ICE: ITS SIGNIFICANCE FOR POLAR ATMOSPHERIC CHEMISTRY

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Polar snow and ice analysis provides potential information to characterize changes in concentrations of important atmospheric trace species such as H_2O_2 and HCHO. In order to evaluate an atmospheric concentration from snow and ice measurements, the transfer mechanisms have to be known. Different steps are involved, starting at the incorporation of the trace species during formation of precipitation in the cloud over processes taking place at the surface of an ice sheet to the processes related to the sintering of snow to firn and ice. For H_2O_2 the different aspects of the transfer mechanism are relatively well known and supported by parallel measurements of atmospheric concentration. For HCHO the information is scarce. An idea of the transfer function has to be gained from a limited amount of pit studies.

Regarding H_2O_2 and especially HCHO, the available results suggest that processes after deposition in the uppermost layer of the firn strongly influence the transfer function.

P1.18

DISTRIBUTION OF DMS AND COS IN THE ANTARCTIC REGION

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Simultaneous shipboard measurements of Dimethyl Sulfide (DMS) and Carbonyl Sulfide (COS) in seawater and the marine boundary layer were performed during a cruise from Punta Arenas (Chile) to Cape Town (South Africa) through the Weddell Sea in November and December 1990. This cruise track was made in connection with measurements carried out from July to November 1990 in the Arctic Sea and the North and South Atlantic.

As DMS is produced biologically in seawater, the concentrations of dissolved DMS showed a reasonable temporal and spatial variability, whereas COS was distributed fairly homogeneous in all the waters examined.

Atmospheric DMS ranged from a few pptv up to 1000 pptv depending on a variety of influencing factors such as concentration in sea water, ice cover and the meteorological situation. Atmospheric COS was characterized by a significant decrease south of 62° southern latitude compared to concentrations found in the northern hemisphere and the southern lower latitude troposphere from July to November 1990.

P5B.4

A NEW DIAL LIDAR FOR MONITORING TROPOSPHERIC AND STRATOSPHERIC OZONE IN ANTARCTICA

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A complex lidar system has been developed under a joint Italian-French cooperative program for measurement of the vertical profile of ozone and of the backscattering and depolarization characteristics of clouds, both in the troposphere and in the stratosphere.

The DIAL system consists of a transmitter with two lasers and a total of five wavelengths and two polarizations. In the transmitting sections, a Nd-YAG laser and an Exciplet laser are mounted. The Nd-YAG laser is provided with a second, third and fourth harmonic generator (532-, 355- and 266-nm wavelengths). The fourth harmonic wavelength pumps two Raman shifters, filled respectively with deuterium and hydrogen. The following wavelengths are obtained: 289 nm and 299 nm. These two lines, both in the ozone absorption band, are used for measurement of the ozone profile between 2000 and 15,000 meters altitude. The 355-nm wavelength may be used for aerosol correction. The two lines are chosen close enough in to neglect the effect of Mie scattering on the measurements. The 532-nm wavelengths is emitted linearly polarized by the Nd-YAG laser and received in two polarization planes in order to measure the backscattering and depolarization characteristics of the aerosol and cloud profiles.

For stratospheric ozone, the 308-nm wavelength emitted by the exciplet laser and the 355-nm wavelength generated by means of the Nd-YAG laser are used for the measurements of stratospheric ozone profiles.

The system has been tested in 1990 at the Observatoire d'Haute Provence and shipped to the French base of Dumont d'Urville where it will start operation at the end of February 1991. The system seems particularly suited for the investigation of stratosphere-troposphere exchange.

P5A.10

INTER-RELATIONSHIPS BETWEEN CLOUD RADIATIVE EFFECTS, TRANSPORT PROCESSES, AND BOUNDARY-LAYER STRUCTURE AT THE SOUTH POLE DURING WINTER

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Radiation and meteorological data collected at the South Pole during the 1986 Austral winter are analyzed to gain a better understanding of the relationships between cloud radiative effects, transport processes and the vertical distribution of temperature and wind. An algorithm is developed to characterize the quasi-permanent surface-based temperature inversion and the warmer, radiatively-active layer above it. Mean winter temperatures and wind profiles for clear and overcast conditions are combined with surface radiation measurements and synoptic circulation patterns to study the mechanisms that cause periodic weakening of the inversion. Results support previous studies that ascribe this weakening to (1) warm air advection, (2) downward vertical mixing of sensible and latent heat, and (3) longwave cloud radiative heating. The integrity of the inversion depends on the combined effects of all three mechanisms. Parameters representing the intensity of the inversion and the bulk wind shear through the lower troposphere are suggested as appropriate indices for the detection of climate in the region of the Antarctic Plateau.

3.4

BIOGENIC BROMINE GASES AT McMURDO: ICE ALGAL PRODUCTION, ATMOSPHERIC CONCENTRATIONS AND POTENTIAL INFLUENCE ON SURFACE OZONE

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Samples of sea ice microalgae were retrieved from McMurdo Sound and incubated at low light and temperature conditions, then sparged through absorbent tubes for later gas chromatography (GC) analysis. It was found that these algae released bromoform, dibromomethane, and mixed bromochloromethanes to the seawater/icewater matrix. Whole air flask samples collected above similar cell suspensions and analyzed by GC-mass spectrometry independently confirmed this finding as well as demonstrating that these compounds were also released to the atmosphere. Algal samples returned frozen to the laboratory were found to contain the same organobromines in body weight concentrations similar to those reported for a variety of macroalgae. Atmospheric measurements at various locations on McMurdo Sound were elevated with respect to air flowing off of the polar plateau, further illustrating that the coastal region is a source of these gases. Some ozone profiles at McMurdo showed that during surface inversion conditions ozone at the surface may be strongly depleted. This is highly reminiscent of the surface ozone depletion events frequently observed throughout the Arctic, which it is believed is initiated by the photolysis of the same organobromine compounds. The amount of ice microalgal biomass in the north and south polar regions is believed to be very large. Using current estimates of this mass and estimates of bromine production rates we calculated that this algal source may be competitive with global macroalgal production and with total anthropogenic production of all organic bromine gases.

P4.2

SEASONAL INPUT OF HEAVY METALS TO ANTARCTIC SNOW

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A sequence of snow blocks was collected at Dolleman Island on the east coast of the Antarctic Peninsula in 1986. The blocks extend to a depth of 1.8 m, and include just over one year's snow accumulation. They were collected with every precaution against contamination, and then sub-sampled in a clean laboratory in the UK to provide a complete sequence of analyses.

The chemistry of the Dolleman Island site is rather well characterized from previous work. The chemical input is dominated by high concentrations of sea salt, peaking in the winter months, and summer peaks of sulfuric acid, presumed to be due to marine biogenic activity.

The snow blocks were analyzed for Cd, Cu, Pb and Zn, as well as for the crustal marker element Al, the marine marker Na, and sulfate. In common with recent data from another site across the Weddell Sea, all Cd measurements were below the detection limit of 0.05 ng kg^{-1}. For Cu and Zn, no clear seasonal variation was apparent. In particular, neither element showed any tendency to vary in parallel with Na. This enables us to place an upper limit on the heavy metal input associated with marine aerosol for these elements.

Although the dataset is rather short, it does appear as if Pb may be varying seasonally, with the peak tentatively assigned to the late winter period. This finding will be discussed in terms of sources and transport of heavy metals and other elements to the region. The heavy metals are an important indicator of long-range transport pathways, and the possible influence of polluted air will be discussed.

P1.5

COMPARATIVE STUDY OF THE BEHAVIOR OF TROPOSPHERIC OZONE IN ANTARCTICA AND IN NORTHERN EUROPE

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The vertical profile has been measured by balloon-borne sondes at the Antarctic Peninsula, Marambio (64°S) in 1988-89 and again since the winter of 1990 continuously as Finnish-Argentine cooperation.

The Finnish Meteorological Institute is responsible for North-European database for ozone soundings and total ozone (three ozone sounding stations, latitudes 67-79°N). Comparisons of the behavior of tropospheric ozone at high southern and northern latitudes will be reported. Seasonal means and correlations between ozone and meteorological factors will be presented. Single anomalies have been studied by calculating three-dimensional trajectories based on the global wind analyses of the ECMWF.

The comparisons show pronounced differences in the troposphere in summer and spring. Tropospheric ozone is highly influenced by anthropogenic NO_x/HC releases at high northern latitudes, whereas at roughly the same southern latitudes, the troposphere is found to be affected mainly by natural (i.e. meteorological) factors.

I.1

CARBON GASES IN THE ANTARCTIC TROPOSPHERE

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The measurements of CO_2, CH_4 and their isotopic ratios over Antarctica define the southernmost limit of the global surface data. The Antarctic data thus play an important role in defining what happens to these gases over the Southern Oceans. Small differences in mixing ratios and isotopic ratios depend on the season, and are only partially understood. Atmospheric mixing is vigorous, so that the "inverse" problem of deriving the intensity of source and sink processes from concentration measurements depends sensitively on the quality of the data.

P2.11

YEAR-ROUND OBSERVATION OF TROPOSPHERIC TRACE CONSTITUENTS AT THE GEORG VON NEUMAYER STATION, ANTARCTICA

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In February 1983 a special air chemistry laboratory was established by the Heidelberg University in the vicinity of the German overwintering station (GVN, 70°S, 8°W), which is situated near the ice edge of the Ekström Ice Shelf. The main objective of the ongoing scientific program is to fill the gap in long-term observations of trace gases and aerosol species between the southern hemisphere oceanic sites and the central Antarctic plateau. The technical design of the autonomous satellite station will be outlined briefly by introducing the entire routine sampling and monitoring programme (i.e. carbon isotopes in CO_2 and CH_4, stable isotopes and ^3H in atmospheric water vapor, particulate chemical and isotopic species, ^{222}Rn, condensation nuclei*, ^{85}Kr*, surface O_3*, and NMHCs*).

In discussing some of the key results emerging from our eight-year observation period, emphasis will be on the chemical, isotopic, and radioactive composition of the Antarctic aerosol. The specific results of carbon isotopic measurements on CO_2 and CH_4 are to be shown in a separate contribution of this conference (I. Levin). The particularly important findings inherent to the long-term and year-round sampled aerosol records are illustrated by:

- A clear annual cycle of the stratosphere/troposphere exchange, which can be derived from the combination of $^7Be/^{210}Pb$ and $^{10}Be/^7Be$ ratios,
- A strong, all year-round persistent influence of open seawater at the GVN site as indicated by the sea-salt record,
- A regular annual cycle of long-range transported mineral dust, showing maximum concentrations in mid austral summer,
- An Antarctic sulfur budget almost exclusively controlled by the marine source of reduced sulfur gases which can be deduced from the annual pattern of nss-sulfate, MSA and $\delta^{34}S$,
- A striking seasonal cycle of total nitrate and $\delta^{15}N$ exhibiting distinct and quite regular maxima between September and November of the nitrate concentration and the $\delta^{15}N$ depletion, respectively.

* Measured by German groups other than Heidelberg University.

P2.7

OZONE DEPLETION AND ITS EFFECT ON UV RADIATION UNDER CLOUDY SKIES IN ANTARCTICA

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Numerical simulations have been made with a combination of an accurate radiative transfer model for a layer-structured plane-parallel atmosphere and the optical parameters of cloudy atmospheres in the LOWTRAN7 code for quantitatively analyzing the effects of ozone depletion and clouds (including cumulus, altostratus, stratus, stratus/stratocumulus, nimbostratus, and cirrus) on solar irradiance at the earth's surface in the UV and visible ranges. This study shows 1) solar irradiance at the surface decreases under cloud conditions as clouds scatter more solar radiation into space and it is very difficult to estimate the UV environment at the surface from satellite measurement data only; 2) the combined effect of cloud scattering and surface reflectance is a major difficulty in estimating solar irradiance at the surface under cloudy skies; 3) because clouds have a larger effect on solar irradiance in the visible range than in UV, the ratios of UV-B (280-320 nm) and UV-A (320-400 nm) to photosynthetic available radiation (PAR, 400-750 nm) are larger under cloudy skies than clear skies if surface albedo is not larger than 0.8; 4) one ratio between ground-based irradiance measurement data at two UV wavelengths is not sufficient to estimate total column ozone and it may result in an error more than 100 D.U. in cases of thick clouds and high reflective surfaces; 5) an empirical method has been developed to estimate total column ozone from ground-based irradiance measurement data at four UV wavelengths under low solar elevation conditions based on accurate radiative transfer simulations and it gives a mean accuracy of 15 D.U. and a maximum error of about 30 D.U. This empirical method will be evaluated with UV spectrometer data from the NSF monitoring station at Palmer Station and from a recent cruise in the Antarctic region.

P5A.6

SOME CHARACTERISTICS OF SOLAR UV RADIATION AT ZHONGSHAN STATION, ANTARCTICA

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Now it is clear that variation of the solar UV radiation does not only affect directly on the biosphere, but also plays an important role in the photochemistry of the troposphere. Photodissociation of nitrogen dioxide by solar UV radiation is a main photochemical process for ozone production in the troposphere. Recently, some theories concerning ozone origin in the troposphere indicate that the ozone amount in the troposphere produced by photochemical processes is larger by 3-4 times than that transported directly from the stratosphere. On the other hand, some observation results show that ozone concentration in the atmosphere near the ground is well correlated to the solar UV radiation at the earth's surface. Therefore, investigation of the solar UV radiation and its variations at the earth's surface has a basic significance for understanding photochemical processes and estimating ozone sources and sinks in the troposphere. For this purpose, observations of solar UV radiation at the earth's surface were begun in several regions in China, such as XiangHe (in the north), GuangZhou (in the south), KunMing (in the southwest), and at ZhongShan station, Antarctica (69°22'24"S, 76°22'40"E).

For better understanding the photochemical processes in the troposphere in Antarctica, some variation characteristics of the solar UV radiation at ZhongShan station are analyzed in this paper based on obtained observation data during June-February 1990.

The main results drawn from data analysis are as follows: In clear sky conditions, the maximum of observed total solar radiation at ZhongShan station is 0.629 kW/m^2; that is lower than 0.744 kW/m^2 observed at USSR Mir station (66°30'S, 90°01'E). Average ratio of solar UV radiation to the total solar radiation is 4.5%. That is comparable to that for the Lasa region. The ratio has an obvious variation during the daytime, from 2.2-2.3% in the morning and evening to 5.8-5.9% near noon. It should be noted that diurnal variation curves of the ratio of UV radiation to the total radiation usually are asymmetric relative to noon. In general, the ratio is larger in the afternoon than in the morning.

On cloudy-sky days, the total solar radiation has a considerable variation during the daytime, but variation of the solar UV radiation is slowed when cloudiness increases from 2 to 8. The portion of the UV radiation in the solar spectrum enlarges from 4.5% to 5.0%, which is close to the 4.8% observed in Lasa region under cloudy conditions.

Varying amounts of low cloudiness may have seriously contributed to solar UV radiation. When low cloudiness is increasing, diurnal variation of solar UV radiation slows down and the ratio of UV radiation to total radiation increases (more than 6.0%). It must be emphasized that composition of the solar spectrum is certainly changed by an increase in low cloudiness. For example, in comparison with a clear sky, when cloudiness increases, a portion of UV radiation in total solar radiation may increase by 32%, while the infrared portion decreases by 26%.

6.7

SOOT IN THE ATMOSPHERE AND SNOW SURFACE OF ANTARCTICA

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Samples of snow collected near the South Pole during January and February 1986 were analyzed for the presence of light-absorbing particles by passing the melted snow through a nuclepore filter. Transmission of light through the filter showed that snow far from the station contains the equivalent of 0.1-0.3 ng of carbon per gram of snow (ng/g). Samples of ambient air were filtered and found to contain about 1-2 ng of carbon per kilogram of air, giving a scavenging ratio of about 150. The snow downwind of the station exhibited a well-defined plume of soot due to the burning of diesel fuel, but even in the center of the plume 1-km downwind, the soot concentration was only 3 ng/g, too small to affect snow albedo significantly. Measurements of snow albedo near large inland stations are therefore probably representative of their surrounding regions.

A similar experiment is planned for Vostok Station in January 1991. Results from that experiment will also be presented.

P1.9

MIXING RATIOS AND SEASONAL CYCLES OF ATMOSPHERIC METHANE AND CARBON DIOXIDE AT PALMER STATION

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Methane and carbon dioxide mixing ratios were measured continuously at Palmer Station, Antarctica, from February, 1982, through November, 1985. Ambient air was transferred from a fixed volume sampling loop to a GC-FID once an hour. Both trace gases exhibited increasing concentration trends from 1982 to 1985. The average rate of increase was 12.6 ppbv/yr for methane and 0.62 ppmv/yr for carbon dioxide. The seasonal amplitude of 30 ppbv for methane was comparable to other Southern Hemispheric observations. Carbon dioxide seasonal variations of 3.4 ppmv were higher than those reported at other sites in Antarctica. Seasonal maxima were recorded for methane and carbon dioxide in the early spring, with minima occurring during late summer in Antarctica. The measurement program will be described, together with an attempt to interpret various data features.

P2.9

RESOLUTION OF PHYSICAL AND CHEMICAL COMPONENTS OF THE 1982 WINTER AEROSOL AT SOUTH POLE STATION BY FACTOR ANALYSIS

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Temporal variations and bivariate correlations among aerosol physical properties and elemental composition have provided insights into transport pathways and chemical transformations between source regions and South Pole Station. In order to obtain further insights we have extended the study by applying multivariate statistical methods for data analysis. Factor analysis, a procedure to explain data variability by linear mixing of components and to compute the composition of the components, has been applied to eight measured variables — aerosol extinction coefficients at four wavelengths (σ_{sp}), condensation nucleus concentration (CN), and elemental concentrations in air of sodium (Na), chloride (Cl), sulfur (S) — in 166 8-hour mean measurements during August and September 1982. Because some variables were highly correlative with each other, we carried out factor analysis in two stages.

First, since σ_{sp} values were among themselves highly correlated at the four wavelengths, the extinction data were re-expressed as two orthogonal factors, one skewed toward short wavelengths and the other toward longer. Then their factor scores were entered into another 6-variable factor analysis with CN and the three elemental concentrations. Na and Cl concentrations, mutually highly correlated and attributed to coarse seasalt aerosol, were assigned to a factor heavily loaded with the longer wavelength extinction factor scores. However, shorter wavelength extinction factor scores, CN, and S concentrations were not well enough correlated to be assigned to a common factor. This new finding indicates that, whereas σ_{sp} is a good predictor of coarse aerosol Na and Cl (seasalt), neither σ_{sp} nor CN are good predictors of aerosol S concentration variability in the winter atmosphere at South Pole Station.

P4.3

UNUSUAL MARINE ION RATIOS IN AN ANTARCTIC PENINSULA ICE CORE

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In most studies of polar snow and ice, it has been generally assumed that the marine aerosol reaches the snow surface with its ionic composition in roughly the same proportions as are found in mean sea water. This then allows the calculation of properties such as "excess sulfate" (or non-sea salt sulfate), which is believed to arise from an acidic component. In recent years, there have been a number of reports from sites around Antarctica of negative excess sulfate values in ice cores. These clearly indicate that the marine Na^+/SO_4^{2-} ratio at least does not remain at sea water values. The Na^+/Cl^- has also been reported to depart significantly from sea water ratios, possibly because of reactions occurring in the atmosphere between sulfuric and sodium chloride.

We now report detailed results from the core drilled at Dolleman Island on the east coast of the Antarctic Peninsula. This site is relatively close to open water, and experiences peaks in sea salt ions during the winter months. However, detailed analysis of the core shows that recurring down the core are sections where very high Na^+ and Cl^- peaks are associated with Mg^{2+} and SO_4^{2-} values well below those expected from sea salt ratios. In some places, the Na^+/Mg^{2+} ratio goes to 10 times the marine value. Mg^{2+} tends to peak on either side of the Na^+ peaks. K^+ and Ca^{2+} behave in a similar fashion to Mg^{2+}, though to a lesser extent.

We have examined a number of possible explanations for the extraordinary ratios we have found. We have a good ionic balance throughout, and believe we have eliminated any possibility of analytical error. Although there is some summer melting at Dolleman Island, leading to the formation of thin ice layers, we cannot explain the pattern we see as a result of the preferential movement of some impurities in percolating water. Diffusion of some components could not produce the pattern we observe. We are therefore left with the intriguing possibility that the ratios must arise either from the existence of unusual ratios in the surface water or on the sea ice near Dolleman Island, or from processes in the atmosphere. Selective freezing of various components may be involved, but we do not yet have a satisfactory explanation.

Although this is clearly an unusual site, it may hold clues to the behavior of the ionic chemistry of much larger areas of ice, where some non-marine ratios have been observed.

P6.3

SEASONAL VARIATION IN ATMOSPHERIC CONTAMINATION AS INDICATED BY ANALYSIS OF A SHALLOW, FIRN CORE FROM LAW DOME, EAST ANTARCTICA

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Results from the measurement of soluble chemical, insoluble particulate and hydrogen peroxide contamination, as well as oxygen isotope ratios in a shallow firn core, drilled to 12.42 m during December 1987 near the summit of Law Dome, East Antarctica, are presented. This core covered eight annual layers to 1979, with at least 20 samples per annual layer being prepared and measured. The indication these results give of atmospheric chemistry and conditions at this site during this period is then discussed. Comparison with data from a nearby automatic weather station and from Casey Station (120 km away) is also discussed.

2.4

SULFATE AND NITRATE AEROSOLS IN THE ANTARCTIC ATMOSPHERE

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Aerosol particles were collected in the Antarctic atmosphere from December 1985 to February 1987 and from February 1988 to January 1989. The degree of acidity of individual sulfate particles was measured by vapor-deposited calcium thin film method. Nitrate composition in particles was identified by vapor-deposited nitron thin film method. Particles were examined by a transmission electron microscope. Size distributions of the sulfate particles were made by sizing about 5000 particles.

Radius of collected particles by an impactor ranges from 0.02 μm to 1.8 μm. The maximum of log radius-number distributions lies at 0.056 μm radius throughout the year. In summer, H_2SO_4 particles were exclusively predominant. The size distribution is in the form of Junge distribution with the slope being -3. Maximum radius of H_2SO_4 particles is 1.8 μm. On the other hand, partially ammoniated sulfate (NH_4HSO_4) particles were present in winter season. Maximum radius of the particles is below 1 μm and number concentration of sulfate particles is very small in winter. These results suggest that the active photochemical formation of H_2SO_4 particles takes place and particles grow up to larger size range in summer. In winter, neutralization of H_2SO_4 particles by NH_3 prevail over acidification of particles by H_2SO_4 condensation. Particles which contain NO_3^- were found in both seasons. They seem to be more frequently present in winter than in summer.

5B.1

TRANSPORT SIMULATIONS OF PASSIVE TRACERS FROM THE NORTHERN HEMISPHERE TO THE SOUTHERN HEMISPHERE AND THE ANTARCTIC REGION

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Atmospheric concentrations of carbon dioxide, halocarbons and other trace gases are increasing mainly due to anthropogenic emission. Though sources of these gases are mainly located in the Northern Hemisphere (NH), the concentrations in the Southern Hemisphere (SH) are also increasing due to atmospheric transport. In the SH troposphere, the concentration of carbon dioxide increases with altitude, opposite to the NH. Large latitudinal gradients of these gases are observed in the equatorial lower atmosphere. The oceanic sink of carbon dioxide in mid-latitudes contributed to the vertical profile. However, atmospheric transport from the NH to the SH is considered to play a crucial role for the distribution of these gases in vertical as well as in latitudinal.

In this paper, the results of two works about transport of passive tracers are presented. In the first work, many long-term trajectories of air parcels are calculated based on observed three-dimensional wind data (NMC data). The distribution of air parcels after six months, which were located in the NH troposphere at the initial time (12Z of January 11, 1987) is examined. In the SH troposphere, numbers of air parcels increase with altitude, because air parcels move upward near the equator and they are transported into the SH through the upper troposphere and the lower stratosphere. Large latitudinal gradient near the equator is also formed.

In the second work, we calculated tracer density using simulated wind data by MRI-Global Spectral Model. In this work, tracers are kept emitting from the surface over four selected regions (Japan, North America, Europe and Amazon). Results from this work accord with those in the first one. A discussion will be made on how tracers from the NH are transported into the SH and the Antarctic region based on the above mentioned works.

P2.8

AEROSOLS OVER THE OCEAN REGIONS

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The continent of Antarctica is very far from anthropogenic sources of aerosols and gases. Since most of the land surface is covered with an ice sheet, natural emission of particles and gases, with the exception of some volcanic sources, will be small. Because of this, studying aerosols over the ocean around the Antarctic continent is very important, as these aerosols will contribute to the source of aerosols over Antarctica by transportation.

Measurements of atmospheric aerosols were carried out during the period from December 1985 to January 1989 over the West Pacific Ocean. An electrostatic aerosol sampler for measuring individual aerosol particle concentrations and a stacked filter unit for collecting fine ($D_p < 2.0$ µm) and coarse ($2.0 < D_p < 15$ µm) particles were used in these observations. These aerosol samples were analyzed by PIXE for 18 elements: Na, K, Ca, Mg, Al, Si, Ti, Cr, Ni, Zn, V, Mn, Fe, Pb, Cl, S, P and Br. Mathematical and chemical methods were applied in these analyses.

The purpose of this paper is to show physical and chemical properties, including concentration, distribution and composition of aerosols over this ocean region, to study long-distance transportation, and to quantify source contribution to marine aerosols.

P1.1

VARIATIONS OF OZONE CONCENTRATIONS IN THE SURFACE LAYER OVER THE WEDDELL SEA

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Results of surface atmospheric and ozone studies over the Weddell Sea from aboard the German *R.V. POLARSTERN* have been presented. Ozone concentration varied in a broad range from 1 ppb to 30 ppb, depending upon air mass origin. An analysis of the data using available synoptic and rawinsonde information has been done. A definite diurnal variation of tropospheric ozone observed during sunny days with low total ozone amount (177 matm cm) may be interpreted as a consequence of the occurrence of photochemical reactions under conditions of increased UV solar radiation.

P1.10

ANTARCTIC TOTAL OZONE AND CARBON MONOXIDE: AN EXPERIMENTAL EVIDENCE OF DISTURBING TROPOSPHERIC CHEMISTRY BY OZONE DEPLETION

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Long-term spectroscopic measurements of total abundances of carbon monoxide and ozone at the Soviet Antarctic base Mirny are presented. During spring months a significant decrease of total ozone was evident for the 1980s. Abundances of carbon monoxide measured during Antarctic summers seem to increase, but a rate of possible trend is not larger than 1% per year. Year-to-year fluctuations of both gases are considerable: for the ozone case, well known quasi-biennial oscillations are evident. The most interesting feature of these variations is their high degree of similarity: positive deviations of January-February CO abundances followed positive deviations of preceding September-November total ozone abundances. Namely, during these months (October-January) seasonal diminishing of CO takes place, according to the generally accepted idea of tropospheric CO being consumed photochemically by hydroxyl. Concentration of tropospheric hydroxyl strongly depends on the level of UV radiation controlled by total ozone. So it is reasonable to explain the above mentioned similarity of CO and ozone variations by the impact of total (mainly stratospheric) ozone abundance on the tropospheric photochemistry through UV radiation. This phenomenon may be important on the global scale in view of possible human-induced decrease of stratospheric ozone.

P1.11

HIGH RANGE-RESOLUTION DIFFERENTIAL ABSORPTION LIDAR SYSTEM DESIGNED FOR MEASURING OZONE IN THE LOWER TROPOSPHERE

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A UV differential absorption lidar (DIAL) system measuring ozone profiles (and eventually measuring ozone fluxes together with a Doppler lidar) in the lower troposphere, which has the potential to be used in Antarctic tropospheric ozone research, is under development in NOAA/Wave Propagation Laboratory (WPL). This lidar system simultaneously transmits laser pulses at 266 nm (online wavelength) and 316 nm (or 299 nm) (offline wavelength) at a repetition rate of 10 pulse per second. A multibeam transmitter design allows near-range coverage and significant signal dynamic range reduction. Numerical simulations show that using this lidar system and a combination of polynomial fitting and Wiener filtering technique for data processing, ozone profiles can be measured with accuracy of better than 5% and range resolution of 50-150 m from the surface to 1.5 km for only 20 laser shots (2 seconds of measurement). With more laser shots (longer integration time), longer detectable range and better range resolution can be obtained.

P2.16

AEROSOL CHEMISTRY AT THE SOUTH POLE 1970-1983

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Samples of atmospheric particulate material were collected in Antarctica beginning in 1970. Initially samples were only collected in the Austral Summer, but by the mid-1970's samples were collected throughout the winter as well. The samples were analyzed by instrumental neutron activation analysis for approximately 40 trace elements. The data from the samples was interpreted as coming from several sources: Crystal weathering, the ocean, volcanoes and anthropogenic activities. The data showed that most of the trace metals measured in Antarctica are from natural sources, and that we must understand the natural system before we can blame things on man's activities. This is particularly true as many natural sources such as volcanism and biogenic processes enrich trace metals by orders of magnitude over their natural levels in the environment.

A major surprise in the research was that we really didn't understand the natural high temperature sources (volcanoes, for example!) as well as we thought. This was shown particularly for the elements selenium and arsenic which are definitely enriched vastly by volcanic processes as well as biogenic ones. In addition, our knowledge about the biogenic sources in the oceans and on land were almost totally unknown, and still needs much work as additional elements are found to be of natural origin every time we look in more detail at the natural system.